

Carbon Dots: A New Type of Carbon-Based Nanomaterial with Wide Applications

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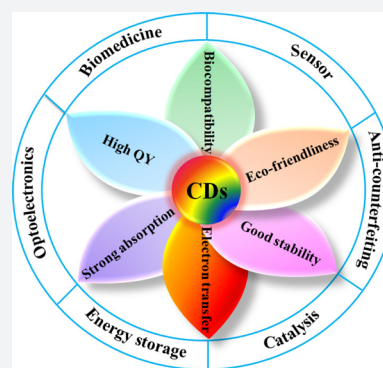
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ABSTRACT: Carbon dots (CDs), as a new type of carbon-based nanomaterial, have attracted broad research interest for years, because of their diverse physicochemical properties and favorable attributes like good biocompatibility, unique optical properties, low cost, ecofriendliness, abundant functional groups (e.g., amino, hydroxyl, carboxyl), high stability, and electron mobility. In this Outlook, we comprehensively summarize the classification of CDs based on the analysis of their formation mechanism, micro-/nanostructure and property features, and describe their synthetic methods and optical properties including strong absorption, photoluminescence, and phosphorescence. Furthermore, the recent significant advances in diverse applications, including optical (sensor, anticounterfeiting), energy (light-emitting diodes, catalysis, photovoltaics, supercapacitors), and promising biomedicine, are systematically highlighted. Finally, we envisage the key issues to be challenged, future research directions, and perspectives to show a full picture of CDs-based materials.



1. INTRODUCTION

Carbon-based materials play significant roles in the development of material science. From the traditional industrial carbon (e.g., activated carbon, carbon black) to new industrial carbon (e.g., carbon fibers, graphite) and new carbon nanomaterials such as graphene and carbon nanotubes (CNTs), fundamental research and applications of carbon-based materials are always popular in the fields of chemistry, materials, and other interdisciplinary due to their environmental friendliness. However, macroscopic carbon material lacks the appropriate band gap, making it difficult to act as an effective fluorescent material. Carbon dots (CDs), a new rising star in the carbon family, have attracted considerable attention due to their excellent and tunable photoluminescence (PL), high quantum yield (QY), low toxicity, small size, appreciable biocompatibility, and abundant low-cost sources, providing important applications in many fields including biomedicine, catalysis, optoelectronic devices, and anticounterfeiting.^{1–9}

CDs can be generally defined as a quasi-0D carbon-based material with a size below 20 nm, and fluorescence is their intrinsic property. In 2004, carbon nanoparticles with fluorescence were first reported, which were accidentally obtained from the purification of single-walled CNTs.¹⁰ In 2006, Sun and co-workers named nanoscale carbon particles synthesized by laser ablation of carbon target as CDs for the first time, but the QY of these surface passivated CDs was only about 10%.¹¹ Low QY and complicated preparation procedures limited the development of CDs. Until 2013, Yang's group chose citric acid (CA) and ethylenediamine as precursors and synthesized polymer-like CDs with QY up to 80% through

one-step hydrothermal method.¹² This QY was the highest value for carbon-based fluorescent materials. These CDs can be applied both as printing inks and as functional nanocomposites. The facile approach, high QY, low toxicity, and high resistance to photobleaching of CDs caused widespread concern and a research boom. Thereafter, researchers developed different strategies and technologies to pursue CDs with high performance, and lots of significant breakthroughs have taken place in the past few years, including multicolor/deep red/near-infrared (NIR) emission,^{2,13–18} narrow full width at half maximum (FWHM),^{2,9,19,20} two-/multiphoton PL,^{2,15,16,21} chirality,^{22,23} room temperature phosphorescence (RTP),^{3,24–27} and thermally activated delayed fluorescence (TADF)²⁸ as well as various applications.

At present, CDs are mainly classified into graphene quantum dots (GQDs), carbon quantum dots (CQDs), and carbonized polymer dots (CPDs) according to their different formation mechanism, micro-/nanostructures, and properties (Figure 1), while associations can be built among them by changing the graphene layer and carbonization degree.²⁹

GQDs possess one- or multiple-layer graphite structures connected chemical groups on the surface/edge or within the

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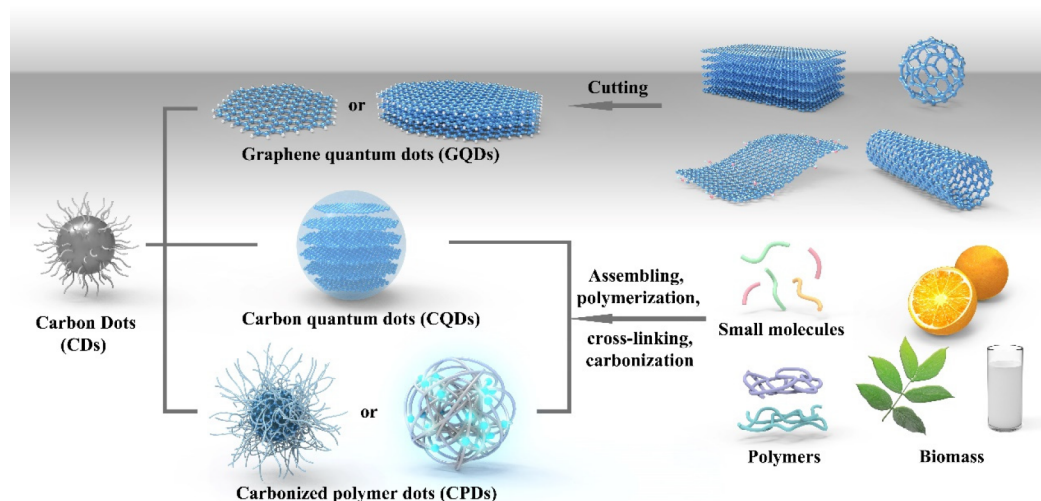


Figure 1. Classification of CDs: including graphene quantum dots (GQDs), carbon quantum dots (CQDs), and carbonized polymer dots (CPDs), and their main preparation approaches.

interlayer defect.^{6,30–35} They have obvious graphene lattices and are usually obtained by “top-down” prepared methods: oxide cutting larger graphitized carbon materials such as graphite powder, carbon rods, carbon fibers, carbon nanotubes, carbon black, or graphene oxide into small pieces.^{30–32,34} Their optical properties are mainly dominated by the size of π -conjugated domains and the surface/edge structures. GQDs are anisotropic with lateral dimensions larger than their height, but CQDs and CPDs are typically spherical, often produced from small molecules, polymers, or biomass by assembling, polymerization, crosslinking, and carbonization via “bottom-up” methods (e.g., combustion, thermal treatment).^{2,16,19,20,24,26} CQDs exhibit multiple-layer graphite structures connected surface groups. Intrinsic state luminescence and the quantum confinement effect of size are their PL mechanism.

Notably, CPDs, showing aggregated/crosslinked and carbonized polymer hybrid nanostructures, were first put forward in 2018 according to the formation process, structures, and PL mechanism.^{19,26} They possess special “core–shell” nanostructures, consisting of carbon cores less than 20 nm with highly dehydrated crosslinking polymer frames or slight graphitization and shells of abundant functional groups/polymer chains,^{2,12,29} which endow CPDs with higher stability, better compatibility, easier modification and functionalization, as well as wider applications. In particular, different from the PL feature and mechanism of GQDs and CQDs, the optical properties of CPDs mainly originate from the molecular state and crosslink enhanced emission (CEE) effect,^{4,6,12,24,27,29,36–39} which make the relationship between structure and performance of CPDs more controllable.

Abundant carbon sources and varieties of synthetic strategies bring the diversity of CDs. Some review articles have summarized the synthesis, PL mechanism, and applications of CDs.^{4–7,29,37,40–43} In this Outlook, aiming at a distinct perspective, we will introduce the excellent optical properties of CDs and highlight the recent significant developments in applications in diverse fields, including optical (sensor, information encryption), energy (catalysis, light-emitting diodes (LEDs), photovoltaics, rechargeable batteries), and promising biomedical applications (bioimaging, phototherapy,

drug/gene delivery, nanomedicine). In the end, we will briefly discuss the current challenges and perspectives of this area in terms of preparation, PL mechanism and application of CDs. We hope this Outlook will offer comparative views and critical insights to inspire more exciting work on CDs for energy, biomedical, and environmental applications in the near future.

2. OPTICAL PROPERTIES

Absorption. CDs prepared from different carbon sources or via different synthetic methods always present different absorption behaviors. However, they typically exhibit strong absorption in the ultraviolet (UV) region (200–400 nm), with a tail extending into the visible range, where the absorption bands are assigned to the π – π^* transition of the C=C bond or the n – π^* transition of the C=O/C=N bond.^{12,44–48} In particular, some CDs with red or NIR emission usually possess π -conjugated electrons in the sp^2 domains and/or the connected surface groups/polymer chains, which results in their long-wavelength absorption in the range 500–800 nm.^{2,49,50} Therefore, the absorption features of CDs are mainly affected by types and content of surface groups, size of π -conjugated domains, and variation of the oxygen/nitrogen content in carbon cores.

Photoluminescence. PL is one of the most appealing properties of CDs from both fundamental and applied perspectives. Compared with other fluorescent materials such as traditional quantum dots (QDs) containing cadmium/lead, rare-earth nanomaterials, and organic dyes, CDs have advantages of better light stability, higher QY, lower toxicity, abundant low-cost sources, and excellent biocompatibility, providing more important applications in diverse fields.

The brightness of PL is quantitatively reflected by the QY value, which is greatly affected by carbon sources, synthetic approaches, and post-passivation. In a broad sense, CDs prepared by “top-down” routes generally possess a relatively low QY compared with “bottom-up” routes.^{2,12,16,19,20,51–53} Thus, the QY of GQDs is always lower than those of CQDs and CPDs. From the perspective of formation and PL mechanism of CDs, more defects formed during the process of oxide cutting carbon resources result in the lower QY of GQDs.^{35,51,53} However, surface modification can reduce the

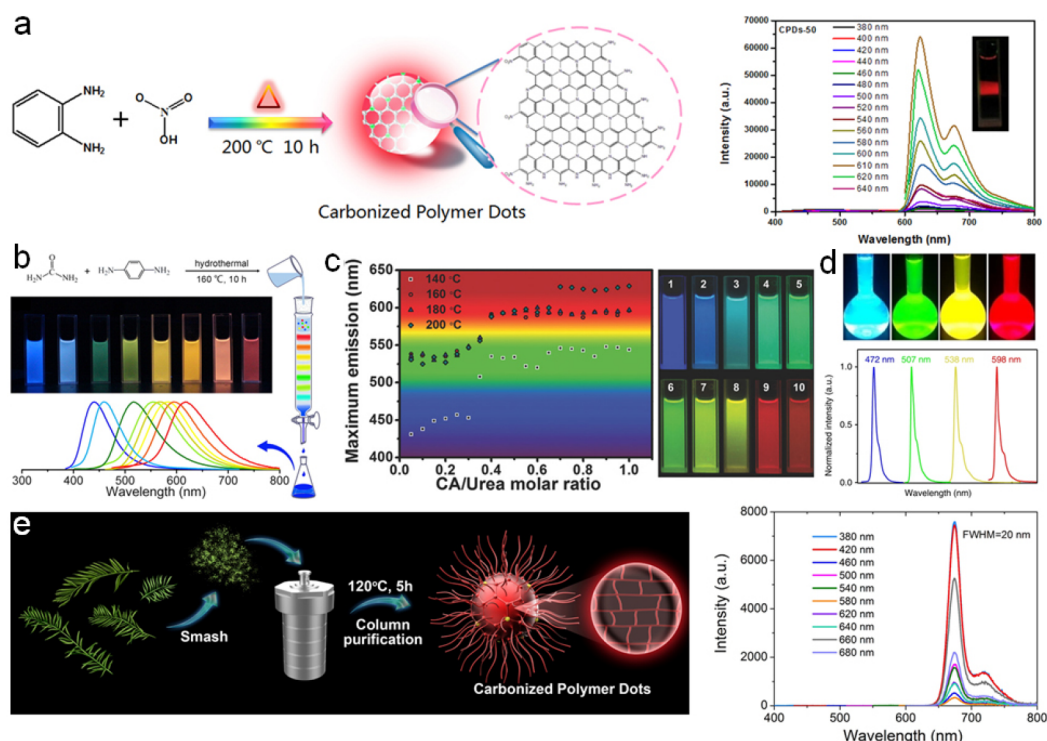


Figure 2. Syntheses and optical properties of CDs. Synthesis and PL spectra of (a) red emissive CPDs¹⁹ and (b) multicolor CPDs.¹³ Optical properties of (c) multicolor CPDs¹⁴ and (d) CQDs.²⁰ (e) Synthesis and PL spectra of deep red emissive CPDs.² Images reproduced with permission from refs 19, 13, 14, 20, and 2. Copyright 2018 WILEY-VCH, 2016 American Chemical Society, 2018 WILEY-VCH, 2018 Springer Nature, and 2020 WILEY-VCH.

nonradiative recombination and enhance the integrity of the π -conjugated system of GQDs, which has been widely used to improve the PL behaviors and regulate the band gaps of GQDs in the past few years.^{6,44,54,55} For CPDs, the CEE effect of crosslinked sub-fluorophores, supramolecular interaction, as well as molecular state emission contribute to their higher QY as compared to completely carbonized CDs (GQDs and CQDs).^{12,56} Now, the QY of 99% has been reported for CPDs in solution.⁵⁶

In most cases, CDs emit blue or green fluorescence, which restricts their further applications in biomedicine, while recent studies have successfully demonstrated the acquisition of red and NIR emissive CDs via adjusting reaction conditions or carbon sources (e.g., aromatic compounds).^{2,13–16,18,19,21,57,58} For example, Yang's group prepared red emissive CPDs with a QY of 31% by modulating the dosage of HNO₃ before the reaction (Figure 2a).¹⁹ Xiong and co-workers synthesized full-color light-emitting CPDs from *p*-phenylenediamine (pPD) and urea (Figure 2b).¹³ In addition to aromatic compounds, by regulating the reaction temperature and ratio of CA and urea, Sun et al. obtained CPDs with multicolor emission (Figure 2c).¹⁴ Cyanine dye and poly(ethylene glycol) were transformed into NIR CPDs with a PL peak at 820 nm through the solvothermal route.⁵⁸ Abundant carbon sources and diverse synthetic methods promote the PL behaviors of CDs.

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Interestingly, there are some differences among the morphology, surface groups, or nanostructures of CPDs, CQDs, or GQDs, while most of them show similar excitation-dependent emission; that is, the PL emission commonly red-shifts with the increase of excitation wavelength. Thus, the PL of CDs can be easily regulated by controlling the excitation wavelength, without changing their chemical structure or size, which is helpful in multicolor bioimaging.^{11,12,48,51,59–61} The excitation-dependent PL behavior and broad PL profile of CDs may come from the multiple PL centers and wide distribution of different energy levels.

On the other hand, researchers have been devoted to CDs with narrow bandwidth emission all the time due to their advantages in bioimaging and optoelectronic devices. Recently, some amazing CDs with FWHM of 20–40 nm have been successfully prepared.^{2,9,20,62} Fan and co-workers reported multicolor excitation-independent emissive CQDs with FWHM of 30 nm (Figure 2d).²⁰ The unique rigid triangular structure, molecular purity, crystalline perfection, and weak electron–phonon interaction of the CQDs surrounded by hydroxy groups result in the high color-purity. Differently, Han et al. found that uniform size distribution leads to CDs with narrow bandwidth emission.⁶² Very recently, the deep red emissive CPDs with unprecedented FWHM of 20 nm and QY up to 59% were prepared from dry taxus leaves, and then purified via silica gel column chromatography (Figure 2e).² Detailed characterizations identified that CPDs had unique polymer characteristics, consisting of carbon cores and the shells of polymer chains, and the efficient π -conjugated system formed with N heterocycles and aromatic rings governs the single PL center, which was responsible for high QY and

narrow FWHM in deep red emissive CPDs. Therefore, the purity, uniform size distribution, single PL center, and simple energy levels play key roles in preparing the narrow emissive CDs.

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Phosphorescence. Room temperature phosphorescence (RTP), one of the attractive properties of CDs, is produced by two critical processes: (i) intersystem crossing (ISC) from the lowest excited singlet state (S_1) to a triplet state (T_n), and (ii) radiative transition from the lowest excited triplet state (T_1) to the ground state (S_0).⁶³ Thus, an effective ISC process and suppressed nonradiative decay are critical for generating RTP from organics. It has been proven that C=O and C=N relevant moieties contribute to the production of RTP because of their strong spin–orbit coupling, causing low singlet–triplet splitting energy.^{42,64} Meanwhile, heteroatoms doping like N, P, and halogens can promote the $n-\pi^*$ transition of C=O and C=N, favoring the ISC process. However, in order to produce RTP, matrices such as poly(vinyl alcohol) (PVA)^{26,65} and filter paper⁶⁴ are used to fix emissive species, stabilize the produced T_1 species, and prevent the CDs triplet states from quenching. Notably, for CPDs, RTP can be easily achieved through appropriate design without additional matrices, because covalently crosslinked frameworks, polymer chains, and supramolecular interactions are beneficial for immobilizing

their emissive centers, further effectively suppressing the nonradiative transitions.^{3,24,25,27} To date, great progress has been made in preparing RTP CDs: from embedding CDs in various matrices to matrix-free CDs as well as expanding applications in multimodal anticounterfeiting and bioimaging.^{3,24,25,27,42,64,65} However, synthesis and applications of CDs-based RTP materials are still at an early stage; their QY, lifetime, and stability should be further improved by designing appropriate structures, post-treatment, or adjusting reaction conditions.

Besides, other luminescent properties of CDs, such as two-/multiphoton fluorescence,^{2,15,16,21,66,67} electrochemiluminescence,^{41,68} and chemiluminescence,^{69,70} have also been successively revealed and explored. Diverse optical properties of CDs lead to their wide applications.

3. OPTICAL APPLICATIONS

Based on their low toxicity, good photostability, aqueous dispersibility, strong PL, and phosphorescence properties, CDs are receiving increasing attention in optical applications including sensor and information encryption, which will be described in this section.

Sensor. CDs have been widely used as fluorescent probes for detecting various analytes in the environment or biological systems due to their intrinsic fluorescent properties, high sensitivity, quick response, low cost, and simple preparation methods. The small size, large specific surface area, and abundant surface functional groups make CDs very reactive and sensitive to the surrounding environment such as temperature, ionic strength, and solvent, resulting in changes to their properties, especially optical properties, for instance, the enhancement/activation (turn-on) and quenching (turn-off) of fluorescence. Theoretically, the detection mechanisms

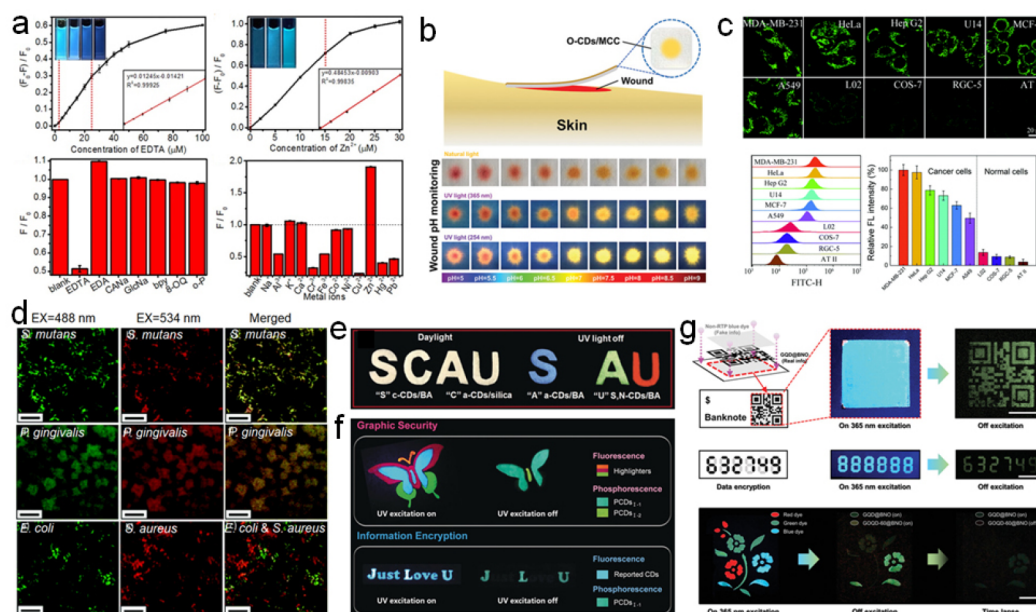


Figure 3. Optical applications of CDs. (a) Zn-doped CPDs were used for EDTA and Zn²⁺ sensing.⁴⁸ (b) Schematic in practical application and images of CPDs combined with medical cotton cloth at different pH values.⁷⁷ (c) Differentiation of various cancerous cells from normal cells by CPDs.⁸⁸ (d) Acidophilic CPDs for distinguishing four bacteria via fluorescent images.⁹⁰ (e) Multicolor RTP CDs for digital information encryption.²⁵ (f) RTP CDs for graphic security and digital information encryption.²⁴ (g) Time-resolved information security using GQDs-based long-lived RTP/TADF.²⁸ Images reproduced with permission from refs 48, 77, 88, 90, 25, 24, and 28. Copyright 2018 American Chemical Society, 2019 WILEY-VCH, 2017 Royal Society of Chemistry, 2019 Royal Society of Chemistry, 2019 WILEY-VCH, 2018 WILEY-VCH, and 2020 WILEY-VCH.

mainly include photo-induced electron transfer (PET), fluorescence resonance energy transfer (FRET), and the inner filter effect (IFE).^{5,71}

CDs are commonly used to detect cations and anions such as Pt^{2+} ,⁷² Hg^{2+} ,⁷³ Zn^{2+} ,⁴⁸ Fe^{3+} ,⁷⁴ ONOO^- ,⁷⁵ and ClO^- ,⁷⁶ which can bind the surface groups of CDs like carboxylate and amino groups through coordination/electrostatic interaction or free radical reaction. Yang's group designed a reversible "off-on" fluorescent nanosensor for selectively detecting ethylenediaminetetraacetic acid (EDTA) and Zn^{2+} through depassivation and repassivation of Zn^{2+} -passivated CPDs, which were prepared from zinc gluconate (Figure 3a).⁴⁸ Interestingly, orange-emissive CPDs with pH-sensitive fluorescence immobilized in medical cotton cloth can be used for wound pH monitoring via both fluorescence and visible colorimetric changes (Figure 3b).⁷⁷ In addition, benefiting from the dual-model response to pH changing as well as the established analytical method, wound pH could be both predicted theoretically and estimated visually in the case of blood contamination and long-term observations. Besides applications in ions and pH sensing, CDs also show selectivity toward a wide range of biomolecules including amino acids (such as cysteine),⁷⁸ glutathione,⁷⁹ vitamins (such as vitamin B₁₂, vitamin B₇, ascorbic acid),^{80–82} formaldehyde,⁸³ glucose,⁸⁴ DNA, and proteins,^{61,85–87} which are associated with health issues and diseases. Therefore, CDs can provide valuable insights into the diagnostics and early precaution of diseases.

Because of the low toxicity, excellent biocompatibility, and photostability, CDs also provide promising probes for efficiently targeting and imaging cancer cells, identifying and detecting bacteria by virtue of their nature or modified ligands. For example, Gao et al. reported mitochondria-targetable CPDs with green emission, which could efficiently distinguish cancer cells from normal cells because of differences in their mitochondrial membrane potentials and substance uptake efficiencies (Figure 3c).⁸⁸ Mao and co-workers designed CPDs–aptamer nanoconjugates for selective detection of living cancer cells.⁸⁹ Yang's group designed acidophilic CPDs displaying blue and red emission to distinguish between four bacteria *Porphyromonas gingivalis* (alkaline environments), *Streptococcus mutans* (acidic environments), *Staphylococcus aureus* (Gram-positive bacteria, cell walls containing teichoic acid), and *Escherichia coli* (Gram-negative bacteria, cell walls lack of teichoic acid) based on different environments and cell walls of these bacteria.⁹⁰ Thus, after labeling with CPDs, compared with *Porphyromonas gingivalis*, *Streptococcus mutans* showed stronger green and red fluorescence and clearer contour, while *Staphylococcus aureus* and *Escherichia coli* only presented one color: bright red and green fluorescence, respectively (Figure 3d). Instead of the traditional indistinguishable bioimaging of these bacteria, this work provided a novel contrast agent to identify bacteria clearly.

Information Encryption. Information encryption is conducive to protecting valuable things from being replicated. Compared with conventional anticounterfeiting technology such as fluorescence printing and plasmonic security labels,^{12,91} or materials including organometallic complexes (metal-containing) and pure organic compounds (metal-free) with RTP, CDs with RTP provide a smart choice for applications in anticounterfeiting data and encryption due to their environmentally friendly feature, easy handling, facile design, and low cost.

Initially, embedding CDs into various matrices such as PVA,^{26,65} filter paper,⁶⁴ and crystalline Mn-framework⁹² was used to obtain CDs-based RTP materials for anticounterfeiting. However, these host matrices only allow specific guest CDs to generate RTP and lack universality. Li et al. reported a universal approach to activate multicolor RTP of CDs in a boric acid matrix for anticounterfeit technology and information encryption (Figure 3e).²⁵ The introduction of B in composites could reduce the energy gap between the singlet and triplet state; facilitating the ISC between S_1 and T_1 , the formed glassy state could protect the excited triplet state of CDs from nonradiative deactivation.

However, this physical immobilization through additional matrices raises challenges, and the instability of reversible interactions in such composites limits their practical applications. To solve the above issues, Yang's group designed novel metal-free RTP CPDs produced from poly(acrylic acid) and ethylenediamine without additional matrices, where amide or imide and the CEE effect were confirmed to favor the generation of RTP.²⁴ In addition, images provided demonstrated the promising applications of CPDs in security protection (Figure 3f). More recently, the transition from RTP to TADF was realized by controlling singlet–triplet energy splitting in GQDs through oxidation.²⁸ The graphics showed that oxidation controlled GQDs systems with both RTP and TADF facilitated anticounterfeiting and multilevel information security (Figure 3g). These researches on structure design and understanding of photophysical properties of CDs offer new directions for developing high-performance afterglow materials.

4. ENERGY APPLICATIONS

Accelerated industrial development and economic growth caused rapid consumption of fossil fuels, energy shortage, environment degradation, and climate change, so exploration of renewable, ecofriendly, stable, and efficient energy conversion and storage technologies is essential and urgent for the global social development.^{7,40,93} As a newly developing carbon material, CDs promise wide applications in the field of energy such as photo-/electrocatalysts, LEDs, supercapacitors, rechargeable batteries, metal–air batteries, and fuel cells, because of their tunable optical properties, low cost, low toxicity, large specific surface area, excellent electron acceptor/donor characteristics, and electronic conductivity.

Catalysis. CDs have been put forward as photocatalysts, electrocatalysts, and photoelectrocatalysts based on their different structures, properties, or purposes.

To make the best of solar energy, a form of renewable and clean energy, photocatalysis has become the noticeable application. CDs are employed as photocatalysts for pollutants degradation (organic dyes,^{94,95} metal ions,^{96–98} etc.), water splitting,^{46,99,100} CO_2 reduction,^{100,101} and chemical reactions¹⁰² in the process of solar energy conversion, which are benefitted from their broad light absorption range, from UV to NIR, low cost, excellent photostability, charge separation, and transfer capability. CDs with nitrogen atoms located at the edge sites of the aromatic domains enable efficient interfacial electron transfer and further lead to an increased photocatalytic hydrogen generation from water.¹⁰³ Usually, CDs hybridized with other nanomaterials (Fe_2O_3 , $g\text{-C}_3\text{N}_4$) serve as the light absorber and/or electron acceptor in the heterojunction to improve the photocatalytic performance.^{97–99,101,104} For example, GQDs-coupled hematite

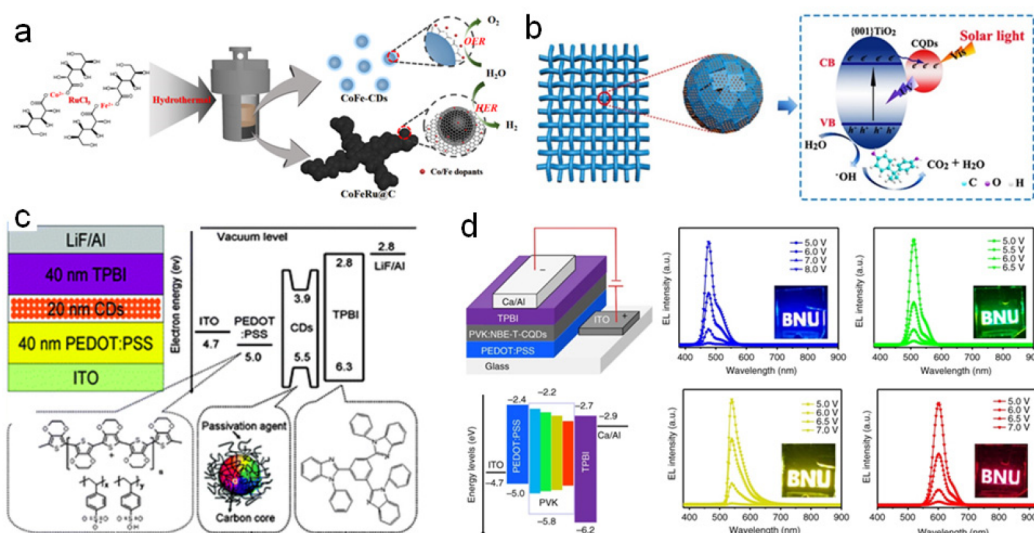


Figure 4. Applications of CDs in catalysis and CLEDs. (a) Illustration of the synthesis of CoFe-CDs/CoFeRu@C catalysts and electrocatalytic OER/HER applications.⁹³ (b) Illustration of the PEC mechanism on the CPDs-{001}TiO₂/Ti photoelectrode.¹¹⁵ (c) Device structure and energy levels of CPDs-based electroluminescent LEDs.¹²⁴ (d) CQDs-based LED structure, energy diagram, and performance characterization.²⁰ Images reproduced with permission from refs 93, 115, 124, and 20. Copyright 2020 Elsevier, 2020 Elsevier, 2011 Royal Society of Chemistry, and 2018 Springer Nature.

exhibited the high photocatalytic H₂ and O₂ evolution rate of 0.390 and 0.225 $\mu\text{mol h}^{-1}$, respectively, which were attributed to GQDs generating a photo-induced potential, enhancing the light absorption of Fe₂O₃, and improving the charge separation efficiency.¹⁰⁵ Yang's group found that the integration of CDs into the g-C₃N₄ backbone facilitated carrier transport in inter-/intra-layers and suppressed recombination, thus dramatically increasing the photocatalytic Rhodamine B degradation efficiency, enhancing photocatalytic rates of CO₂ reduction and water splitting for H₂ evolution, which make it possible to couple environmental remediation with energy conversion.⁹⁵

The oxygen reduction reaction (ORR), oxygen evolution reaction (OER), and hydrogen evolution reaction (HER) are the important energy conversion processes between chemical energy and electric power. Considering low cost, chemical inertness, large specific surface area, high electron mobility, abundant surface defects, and active sites, CDs have become the strong competitor for the conventional Pt- and Ir/Ru-based electrocatalysts. For pure CDs electrocatalysts, heteroatom doping (B, P, S, and/or N) is used to modulate electrical structures and electron-coupling interactions, thus enhancing their electrocatalytic performance.^{4,7,8} Specifically, dual-doped CDs always show a higher ORR catalytic activity than non-doped or single-doped CDs due to higher heteroatom loading, synergistic electron-coupling interactions, and more active sites.^{106,107} However, usually, CDs are combined with other carbon materials (e.g., graphene, reduced GO),¹⁰⁸ some metals (e.g., Ag, Au, Ni, Pd, Fe, Ru),^{93,109–111} or metal-based semiconductors (e.g., Bi₂O₃, Co₃O₄, MoS₂)^{104,112,113} to improve durability and synergistically enhance catalytic activity. For instance, GQDs greatly boosted both OER and HER performance of NiCo₂P₂ by controlling catalyst morphology, enhancing charge transfer, and improving the catalytic kinetics.¹⁰⁹

Actually, the complicated synthesis procedure and high-temperature calcination are contradictory to energy saving and emission reduction. Recently, Yang's group simultaneously synthesized Co and Fe co-doped CDs (CoFe-CDs) with OER

activity and carbon-loaded Co and Fe co-doped ruthenium nanoparticle networks (CoFeRu@C) with HER activity within a one-pot hydrothermal reactor under mild conditions (Figure 4a).⁹³ Benefitting from the synergistic effect of bimetallic dopants and large specific surface area of CDs, the electrocatalytic performance and durability of CoFeRu@C and CoFe-CDs were both superior to the commercial Pt/C and RuO₂, respectively. Remarkably, some CDs can electrocatalyze CO₂ reduction into alternative liquid fuels (e.g., CO, CH₄) or value-added organic chemicals, which has attracted extensive research interest.^{104,112,113}

In the processes of photoelectrochemical (PEC) water splitting and wastewater treatment, CDs act both as photosensitizers to capture more solar energy and electron media to promote charge separation in semiconductors. For example, CPDs-modified anatase/rutile TiO₂ worked as the photoanode for PEC water splitting with a much increased efficiency, because of CPDs simultaneously enhancing the OER kinetics and UV–vis light harvesting.¹¹⁴ Furthermore, utilizing a superior up-conversion performance and enhanced hole–electron separation, the introduction of CPDs could greatly improve the PEC activity toward organic pollutants of {001}TiO₂/Ti (Figure 4b).¹¹⁵ However, the clear working mechanism of CDs-based catalysts is still under investigation, and much effort needs to be devoted to making a breakthrough in the near future.

CDs-Based Light-Emitting Diodes (CLEDs). LEDs, solid-state lighting devices for converting electric energy into light, have been a hotspot in academic research for years, aiming at their applications in liquid-crystal displays, full-color displays, and illumination devices in our daily life. As a newly developing fluorescent material, CDs are expected to replace expensive rare-earth-based phosphors and toxic metal-based semiconductor QDs in LEDs, because of their abundance, adjustable PL colors, low cost, and environmental friendliness. Generally, CDs can act as phosphors in CLEDs or active layers in electroluminescent devices.

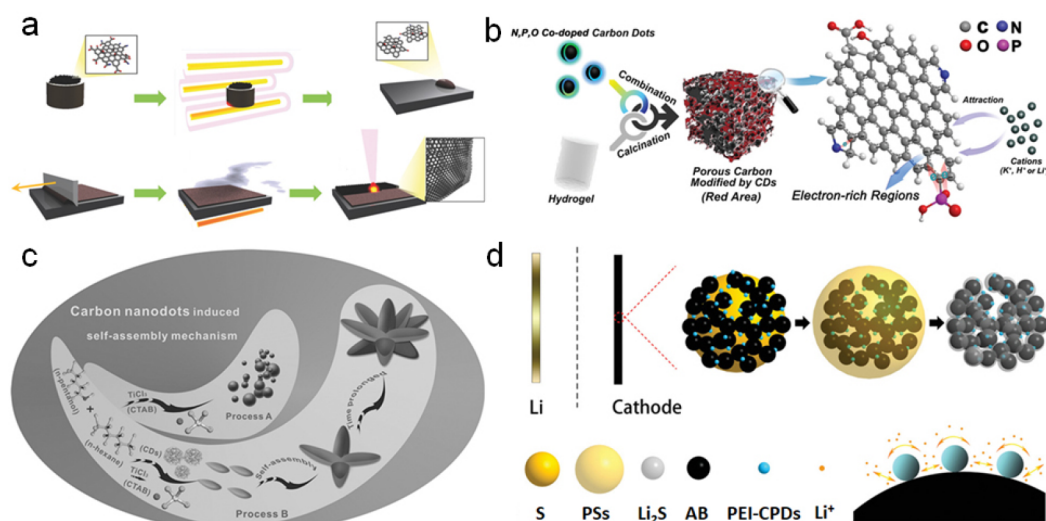


Figure 5. Applications of CDs in supercapacitors and rechargeable batteries. (a) Illustration of conversion process.¹³³ (b) Porous carbon derived from CPDs and polyacrylamide hydrogels.¹³¹ (c) Schematic representation of the formations of graphene-rich petal-like rutile TiO₂.¹³⁴ (d) Illustrations of the PEI-CPDs-modified cathode composite at different charge-discharge stages.¹⁴⁰ Images reproduced with permission from refs 133, 131, 134, and 140. Copyright 2018 WILEY-VCH, 2019 WILEY-VCH, 2016 WILEY-VCH, and 2019 WILEY-VCH.

In phosphor-based CLEDs, most CDs are homogeneously dispersed in matrix materials such as transparent polymers (e.g., PVA,^{38,116} polystyrene,¹¹⁷ epoxy resins¹¹⁸), starch,¹⁷ silica,¹¹⁹ or even inorganic salts,¹²⁰ to prevent aggregation-caused quenching of CDs in the solid-/powder-state resulting from excessive resonance energy transfer or direct π – π interactions. Multicolor/white CLEDs have been demonstrated through these methods. However, these common methods need multistep procedures, and the introduction of a thick solid matrix layer can depress the luminescence from the chip and increase the cost. In recent years, some self-quenching-resistant solid-state fluorescent CDs were developed from polymers,¹²¹ inorganic salts,¹²² or silane coupling agents¹²³ and directly used as phosphors in CLEDs, which greatly simplified the preparation process and reduced the cost. Nevertheless, up to now, there are still challenges for obtaining multicolor/white CLEDs with high luminous efficiency and color rendering index (CRI). More attention should be paid to developing high-performance CDs with solid-state fluorescence, high QY, and narrow FWHM.

For electroluminescent CLEDs, CDs usually serve as an active emission layer sandwiched in the multilayer device structure. Ma and co-workers first demonstrated a white CLEDs device by using CA-derived CPDs as the emissive layer, of which the structure and energy level diagram are shown in Figure 4c.¹²⁴ A maximum luminance (L_{\max}) and an external quantum efficiency (EQE) of 35 cd m^{−2} and 0.083% were realized, respectively. Inspired by this work, some great progress has been made recently for electroluminescent CLEDs applications. For instance, Yuan et al. employed high-performance multicolor triangular CQDs as emissive layers to fabricate multicolor CLEDs, which presented high color-purity (FWHM of 30–39 nm), L_{\max} of 1882–4762 cd m^{−2}, and current efficiency of 1.22–5.11 cd A^{−1} (Figure 4d).²⁰ Subsequently, controlling the device fabrication and modification/passivation of CDs, they reported deep-blue CLEDs with L_{\max} exceeding 5000 cd m^{−2} and EQE of 4%, which substantially outperformed deep-blue LEDs based on Cd²⁺/Pb²⁺-comprising materials.⁹ Nevertheless, the research of

CLEDs is still at an early stage, compared with well-developed Cd²⁺-QDs based LEDs (QLEDs) or excellent perovskite QLEDs; CLEDs show definite room for improvement in luminance, EQE, and CRI.

Solar Cells. CDs have been extensively studied in solar cells (SCs), where a small amount added to the bulk heterojunction has been found to improve efficiency in various works due to their unique optical properties, abundant functional groups (e.g., amino, hydroxyl, carboxyl), and high electron mobility. The roles of CDs playing in SCs mainly include (i) facilitating separation of photogenerated electron–hole pairs, suppressing carrier recombination; (ii) broadening the light absorption range; and (iii) improving device environmental stability. Like CPDs-based perovskite SCs, CPDs can dramatically enhance long-term stability against high humidity and high temperature, convert the harmful UV light into desirable visible light, and promote carrier separation and electron extraction for improving cell efficiency.^{57,125,126} Additionally, Zhang et al. optimized the performance of GQDs/Si heterojunction SCs by controlling the size of GQDs as well as thickness of the GQDs layer, which impacted the hole transport and carrier recombination.¹²⁷

Supercapacitors. Supercapacitors show fast charge/discharge rates, high power density, and long cycle life but inferior energy density, which limits their practical application in energy storage. CDs hybridized with other carbon materials,^{128,129} polymers,^{130,131} or metal oxides^{129,132} are reported to improve the electrochemical performance of supercapacitors.

CPDs possess continuously interconnected core–shell structures, crosslinked networks, and abundant functional groups, which facilitate to form a continuous conductive 3D carbon skeleton with large specific surface area and good wettability, and fabricate the stable and flexible multilayer energy storage devices. Kaner et al. converted N and O co-doped CPDs into open porous 3D turbostratic graphene networks to fabricate supercapacitors (Figure 5a), finally obtaining fast charging rates with a relaxation time of 3.44 ms and energy density of 17.7 W h kg^{−1}, which was much

higher than the commercial aluminum-based capacitor.¹³³ By designing hierarchical porous carbon (HPC) materials from N, P, O co-doped CPDs and polyacrylamide hydrogel (Figure 5b), Xiong's group realized supercapacitors with energy densities of 62.8–90.1 W h kg⁻¹.¹³¹ This demonstrated that N, P, O codoped CPDs not only reduced interface resistance of porous carbon materials but also enhanced conductive stability of the electrode at high scan rates. QDs can also enhance the capacitance of activated carbon¹²⁹ or MnO₂ nanosheets¹³² due to their unique conductivity and high crystallinity.

CPDs possess continuously interconnected core–shell structures, crosslinked networks, and abundant functional groups, which facilitate to form a continuous conductive 3D carbon skeleton with large specific surface area and good wettability, and fabricate the stable and flexible multilayer energy storage devices.

Rechargeable Batteries. Rechargeable batteries are considered as one of the most effective energy storage technologies to bridge production and consumption of renewable energy. In Li, Na, or K ion batteries, CDs can create an excellent interface for intercalations between electrodes and electrolytes via the surface engineering, provide more active sites for ion insertion and extraction, help to boost the stability, enhance electron/ion transfer and diffusion, and improve electrochemical properties.^{134–139} CPDs have been used as “designer additives” for graphene-rich petal-like rutile TiO₂ to shorten diffusion pathways of Na⁺ and increase conductivity of the ensemble, which resulted in sodium storage with high capacity and long-term cycle stability (Figure 5c).¹³⁴ Besides, oxygen-containing functional groups from CDs facilitate to attract metal cations, produce a uniform solid electrolyte interphase, and maintain the integrity of the electrode structure, thus improving electrochemical performance.¹³⁵ To alleviate the effect of the polysulfide shuttle, raise the areal sulfur loading, and increase the operation current density in lithium–sulfur (Li–S) batteries, Xiong et al. designed the polyethylenimine-functionalized CPDs (PEI-CPDs)-modified cathode (Figure 5d).¹⁴⁰ This demonstrated that the small size, excellent dispersibility, and greater number of amine groups of PEI-CPDs provided abundant absorption sites that could effectively suppress the polysulfides' dissolution and increase the Li⁺ conductivity around the solid–electrolyte interface.

5. BIOMEDICAL APPLICATIONS

It is not surprising that one of the most promising and frequently reported applications of CDs is in biomedicine. *In vitro* cytotoxicity studies on a series of cell lines demonstrate the low toxicity or nontoxicity and excellent biocompatibility of CDs even at a high concentration level.^{2,12,52,60,141,142} *In vivo* experiments indicate that CDs are rapidly excreted via the

kidney and/or hepatobiliary system. Besides, no remarkable symptoms of inflammation are observed in the brain, heart, lung, liver, spleen, kidney, testicle, and bladder in rats based on blood biochemistry and hematological analysis.^{2,19} Thus, CDs are safe for biomedical applications. In addition, low cost, small size, controllable surface functions, high photostability, unique down-conversion PL, multiphoton PL, and high brightness make CDs promising alternative photo-/nanomaterials to traditional fluorescent materials in disease diagnosis, therapy, and healthcare supplements. In this section, biomedical applications of CDs including bioimaging, phototherapy, drug/gene delivery, and nanomedicine are described.

Bioimaging. Bioimaging is a technique that can directly visualize biological events in real-time and non-invasive ways via probes and detectors. Fluorescence imaging, one of the imaging modalities, has become a powerful approach for clinical diagnosis due to its convenience, low cost, high sensitivity, noninvasiveness, and long-term observation. However, conventional fluorophores like QDs and organic dyes suffer from either toxicity concerns or poor fluorescence performance, which provoke the development of CDs. High photostability, excellent biocompatibility, simple synthetic routes, flexible designability, multicolor emission, deep red/NIR emission, and two-/multiphoton PL make CDs the next-generation fluorescent probes for both *in vitro* and *in vivo* bioimaging.

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Various CDs have been used extensively for imaging cells, microorganisms,^{90,143–148} and plant tissue.¹⁴⁹ Generally, CDs can quickly enter into cells through energy-/temperature-dependent macropinocytosis-, clathrin-, caveolae-, and/or lipid raft-mediated endocytosis and are distributed into mitochondria,^{45,150,151} lysosomes,^{19,142,152} endoplasmic reticulum,^{19,142,152} Golgi apparatus,^{153,154} and/or nucleolus^{155,156} based on the different nanostructures of CDs and types of cells. Imaging organelles (e.g., imaging mitochondria and/or nucleolus) is profitable to understand and study organelle-related diseases such as cancer, Alzheimer's disease, Parkinson's disease, diabetes, and cardiac dysfunction. Wu et al. found that CPDs prepared from mPD and L-cysteine can realize nucleus-targeted imaging in both fixed cells and living cells (Figure 6a).¹⁵⁵ Furthermore, conjugated with protoporphyrin IX, CPDs obtained a nucleus-targeted PDT ability, which caused effective tumor ablation without toxicity effects after laser irradiation. *Lactobacillus plantarum* derived CPDs served as staining agents for imaging biofilm-encased microorganisms (Figure 6b), which could give valuable information about the morphology and physiological state of bacteria in a biofilm.¹⁴⁵ Additionally, *L. plantarum* derived CPDs displayed better photostability than commercial dye SYTO 9, and this

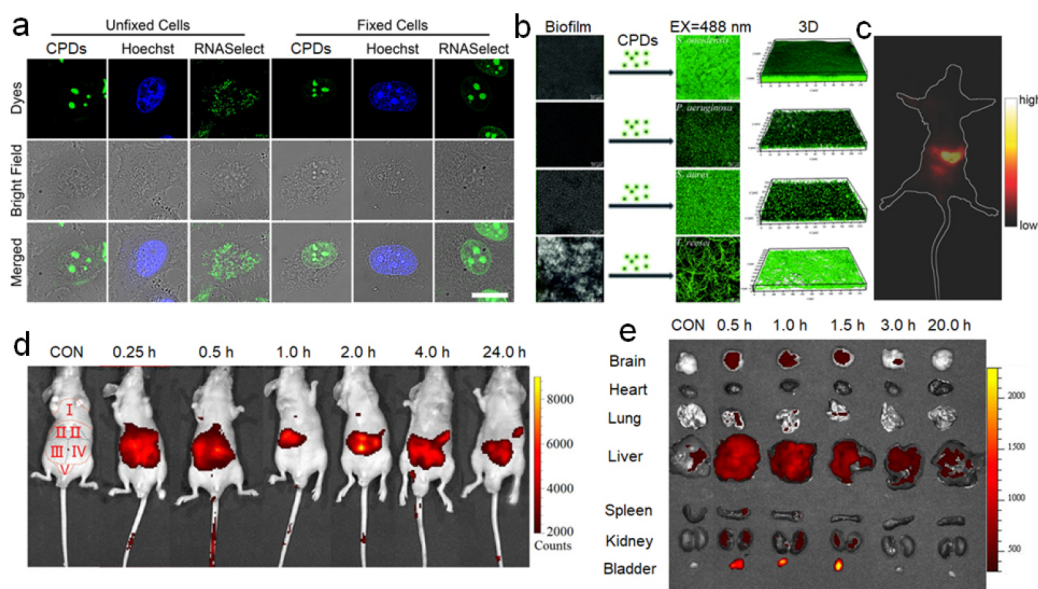


Figure 6. Application of CDs in bioimaging. (a) Confocal images of unfixed and fixed HeLa cells stained by CPDs, Hoechst, or SYTO RNaselect.¹⁵⁵ (b) Biofilms of different microorganism species stained by CPDs.¹⁴⁵ (c) Deep red emissive CPDs for stomach imaging.¹⁵ (d) *In vivo* imaging of nude mice with intravenous injection of deep red emissive CPDs at different time points.² (e) Real-time *ex vivo* imaging of nude mice with intravenous injection of red emissive CPDs in phosphate buffer saline solution at different time points.¹⁹ Images reproduced with permission from refs 155, 145, 15, 2, and 19. Copyright 2018 American Chemical Society, 2017 Royal Society of Chemistry, 2018 WILEY-VCH, 2020 WILEY-VCH, and 2018 WILEY-VCH.

imaging method was much easier and universal as compared to fluorescent proteins as probes.

Apart from *in vitro* imaging, benefiting from minimum autofluorescence and light scattering by tissues, offering great imaging contrast and spatial resolution, CDs with red/NIR emission or two-/multiphoton PL have been excellent candidates for *in vivo* fluorescence tracking agents. Sun et al. first reported the *in vivo* imaging of CDs by adopting three routes in mice.¹⁵⁷ In addition, the clearance rate of CDs was ranked as intravenous (tail vein), intramuscular (muscle of left leg), and subcutaneous (under the skin of left leg).⁴⁷ Qu and co-workers developed two- and three-photon-induced deep red emissive CPDs through surface engineering and applied them for *in vivo* deep red fluorescence imaging of the stomach of a living mouse (Figure 6c).¹⁵ Recently, Yang's group reported deep red emissive CPDs with QY up to 59% as an efficient probe for both one-photon and two-photon bioimaging.² Besides, they systematically investigated the biodistribution of CPDs in nude mice via real-time *in vivo* imaging (Figure 6d). Results demonstrated that CPDs could rapidly enter the whole body of mice with blood circulation in a few minutes. Most CPDs first heavily accumulated in the liver, lung, and kidney and then were gradually cleared via both the kidney and hepatobiliary system within 24 h. Excitingly, they found that red emissive CPDs prepared from oPD could easily cross the blood brain barrier of healthy mice without targeting agents (e.g., transferrin) (Figure 6e), which provided a new material for the prevention and theranostics of brain diseases via real-time tracking.¹⁹

The fluorescence imaging technique of CDs shows a high contrast ratio and high sensitivity, but the spatial resolution is still unsatisfactory in biology and clinic applications because of the limited penetration depth. Thus, there is an emerging need to develop multimodal imaging probes through the combination of other imaging modalities efficiently, for instance, photoacoustic (PA) imaging, magnetic resonance (MR)

imaging, and computed tomography (CT) imaging. PA imaging, as a hybrid imaging modality, has advantages of both optical and ultrasound for deep tissue penetration as well as high spatial resolution. CDs, as PA contrast agents, should have high absorbance coefficients in the red to NIR region and the effective ability to convert light into heat.¹⁵⁸ In this respect, red/NIR emitting CDs or CD composites are always exploited as a fluorescence and PA bimodal probe for *in vivo* imaging.^{158–160} Other multimodal CDs can be obtained by doping MRI/CT probes (e.g., Gd,¹⁶¹ Mn,¹⁶² Yb)¹⁴¹ into CDs. These researches identify that CDs exhibit an excellent multimodal imaging performance, which can enhance the efficacy of imaging-guided theranostics through offering visual guidance and information, for example, agent location and circulation in physiological environments, which will be discussed in the following sections.

Phototherapy. Phototherapy, including photodynamic therapy (PDT) and photothermal therapy (PTT), is a form of noninvasive therapeutic treatment that converts the irradiating light into reactive oxygen species (e.g., $\bullet\text{OH}$, $\text{O}_2^{\bullet-}$, and $^1\text{O}_2$) and heat with the help of photosensitizers, inducing local apoptosis of cancer cells. CDs have gained much attention as promising phototherapeutic agents due to their unique optical properties, high water solubility, and high photostability.

It is reported that the hypoxic tumor microenvironment and rapid consumption of oxygen in the PDT process can severely impede therapeutic effects of CDs because of the oxygen-dependent PDT. Besides, worsened hypoxia, caused by PDT-induced oxygen consumption, would result in irreversible tumor metastasis or drug resistance. To overcome the hypoxia of the tumor tissue, Zhang et al. designed CPDs-decorated C_3N_4 nanocomposites for light-driven water splitting to improve the intratumoral oxygen level and ultimately reverse the hypoxia-triggered PDT resistance and tumor metastasis (Figure 7a).¹⁶³ The Mn-doped CPDs assembly also could

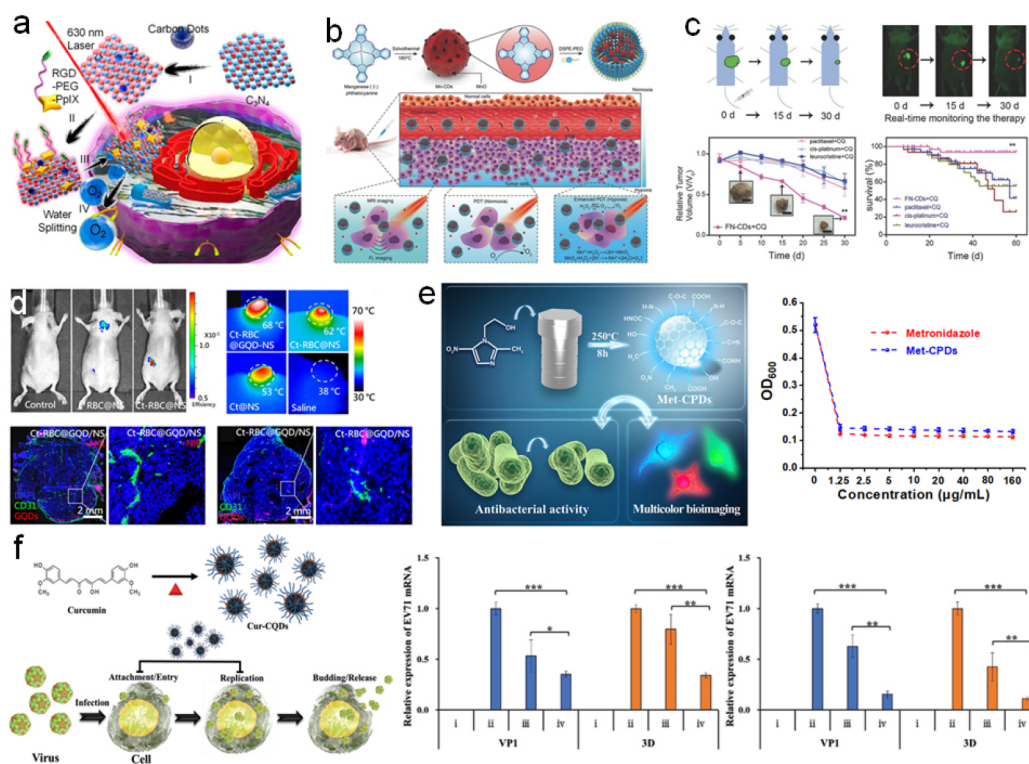


Figure 7. Biomedical applications of CDs. (a) Structure of CPDs-decorated C_3N_4 nanocomposites and schematic diagram of 630 nm light-driven water splitting-enhanced PDT.¹⁶³ (b) Schematic illustration of the Mn-CPDs assembly for enhanced PDT.¹⁶⁴ (c) Effectiveness and monitoring of the tumor-targeted therapy based on FA-CPDs *in vivo*.¹⁷³ (d) *In vivo* study of GQDs-based nanomaterials with A549 tumor-bearing BALB/c nude mice.¹⁷⁴ (e) Illustration of the synthesis of Met-CPDs and their antibacterial activity against *Porphyromonas gingivalis*.⁶⁰ (f) Illustration of the synthesis of Cur-CPDs and their antiviral applications.¹⁸⁶ Images reproduced with permission from refs 163, 164, 173, 174, 60, and 186. Copyright 2016 American Chemical Society, 2018 WILEY-VCH, 2018 WILEY-VCH, 2019 American Chemical Society, 2017 Royal Society of Chemistry, and 2019 WILEY-VCH.

effectively generate 1O_2 (QY of 0.40) and catalyze H_2O_2 to produce oxygen under hypoxia environments further for simultaneous bimodal fluorescence/MR imaging and enhanced PDT (Figure 7b).¹⁶⁴ In addition, molecular dyes (e.g., porphyrin,¹⁶⁵ diketopyrrolopyrrole¹⁶⁶), Ru,⁶⁶ or Mn-function-alized¹⁶⁷ CDs were used for improving the PDT efficacy (e.g., QY, 1O_2 yield) and/or multimodal imaging-guided PDT *in vivo*.

Red/NIR emitting CPDs prepared from CA and urea,^{18,168,169} polythiophene, and diphenyl diselenide⁶⁷ were recognized as effective theranostic agents for PTT with high photothermal conversion efficiency in excess of 50% due to their unique red/NIR absorption feature, which were superior to that of conventional photothermal agents (such as Au-based nanostructures).¹⁷⁰ Nevertheless, PTT usually requires high-power and long-time laser irradiation to produce adequate heat to kill cells, and the up-regulated expression of heat shock proteins greatly decreases the treatment effect.¹⁷¹ To avoid drawbacks of the mono-mode therapy, synergistic PDT and PTT are adopted to cancer therapy.^{159,171,172} Taken together, these works extend the application of CDs in the cancer therapy field.

Drug/Gene Delivery. Aside from anticancer phototherapies, CDs can combine imaging tools with drugs or genes to form imaging-guided nanohybrids for improving the delivery efficiency or offering benefits in the therapeutic strategy.

Drug delivery, the safe and efficient treatment, refers to carrying the medicine to a specific location in the body and

releasing it in a sustained manner. Thus, the controlled drug release and robust selectivity in drug delivery systems are crucial for increasing local therapeutic effects and minimizing side effects of non-infectious and/or non-cancerous tissue. CDs have advantages in visualizing drug accumulation and activities at pathological sites via their fluorescent properties, which are vital for estimating therapeutic efficacies of medicines.^{173–177} Through tracking the green emission from folic acid-modified CPDs (FA-CPDs) combined with chloroquine, Fan et al. realized the real-time imaging/monitoring of tumor therapy (Figure 7c).¹⁷³ Similarly, the encapsulation of GQDs/anticancer drug docetaxel in a nanosponge demonstrated effective drug delivery, imaging, and photolytic abilities against deep tumors (Figure 7d).¹⁷⁴ To improve tumor-specific imaging and drug delivery performance, Zhou and co-workers designed deep red emissive CQDs with multiple paired α -carboxyl and amino groups, which could target tumors including glioma due to their multivalent interactions with large neutral amino acid transporter 1. Thus, loaded with topotecan hydrochloride, the CQDs could be used for fluorescence/PA imaging and the treatment of brain cancer, showing potential clinical applications in imaging and drug delivery diseases of the central nervous system.¹⁷⁵

Different from other therapy technologies, gene therapy is considered as a durable and possibly curative clinical strategy for various diseases such as inherited human diseases and cancers.^{178–180} Effective vectors in gene therapy can deliver genetic materials into cells, and possess high gene transfection efficiency. Viral vectors with the natural ability to invade and

deliver their genetic material have served as effective gene carriers. However, severe safety risks based upon their immunogenicity and their oncogenic potential have kept them far from safe for clinical use.¹⁷⁹ CDs possess low toxicity, abundant functional groups, and excellent biocompatibility. Importantly, the small size of CDs contributes to adequate cellular uptake of vectors, enhancing gene transfection efficiency. In addition, their unique fluorescence can be used for the tracking the internalization of genes.^{178,179} Thus, CDs will be attractive as non-viral vectors in gene therapy.

Nanomedicine. Besides being carriers, CDs themselves behave with therapeutic performances such as antibacterial activity,^{60,181–183} anticancer activity,^{184,185} antiviral activity,^{186,187} and antioxidant activity.^{188–190}

Usually, using drug molecules (e.g., metronidazole,⁶⁰ gentamicin sulfate,¹⁸¹ glycyrrhizic acid¹⁸⁷) as precursors, the as-prepared CPDs possess similar or superior therapeutic performances as compared to pristine drugs due to the retention of pharmacophores in their structures or the formation of new active structures. More importantly, compared with drug molecules, these drug-CPDs show better biocompatibility and water solubility as well as stronger fluorescence and can be used as efficient bioimaging probes for theranostics. For instance, Yang's group prepared Met-CPDs by hydrothermal treatment of metronidazole, a wide-spectrum antibiotic against obligate anaerobes (Figure 7e).⁶⁰ Compared with metronidazole, Met-CPDs behaved with better aqueous solubility and excellent biocompatibility because of the formation of new functional groups like carboxyl, hydroxyl, and amino groups. Besides, biological experimental data demonstrated that the Met-CPDs showed excellent selective antibacterial activity against obligate anaerobes due to the contained nitro group, a pharmacophore, which was in accordance with the main mode of action of metronidazole.

Differently, Huang and co-workers found that Cur-CPDs derived from curcumin (natural compound with antimicrobial, anticancer, anti-inflammatory, and antioxidant characteristics) were efficient antiviral agents against enterovirus 71 (EV71) (Figure 7f), but the precursor curcumin had insignificant inhibitory activity against EV71 infection in RD cells.¹⁸⁶ This is because the distinctively pyrolytic curcumin polymers on the CPDs contributed to the excellent aqueous solubility and antiviral activity of Cur-CQDs against EV71 and their remarkable biocompatibility. However, further studies are still needed to clearly identify the exact molecular mechanism of these drug-CPDs in the antibacterial, anticancer, antiviral activities.

6. CURRENT CHALLENGES AND OUTLOOK

Facile, environmentally friendly, and various synthetic methods, excellent optical and electrical properties, low cost, as well as good biocompatibility make CDs popular in optical, energy, and biomedical fields. Great progress has been made in the study of CDs including synthetic strategies, structures, properties, mechanistic investigations, and application development. These encouraging research results demonstrate that CDs can provide numerous special opportunities to investigate new phenomena and properties observed in multidisciplinary circumstances, though there are many critical issues yet to be addressed.

Compared with QDs and other carbon materials, research on CDs is still in a more nascent stage. At present, one of the most fundamental and important issues is the lack of a

systematic and scalable synthesis protocol to produce high-quality CDs with desirable structures (e.g., size, shape, crystallinity, numbers of functional groups, type and location of defects). Additionally, their exact reaction mechanism, nucleation mechanism, and formation process are also unclear due to non-standard synthetic pathways and impurities. Therefore, for large-scale production of CDs with high performance through an efficient route, effects of precursors and reaction conditions (e.g., temperature, time, pH) on the performance of CDs should be systematically explored, and a purification scheme based on size or polarity also needs to be developed. Notably, developing *in situ* techniques is necessary for characterizing the formation mechanism of CDs, which contributes to controllable syntheses of CDs with definite nanostructures.

Currently, the PL mechanism of CDs remains controversial due to the different reaction conditions and precursors. It is reported that their PL behaviors are mainly determined by structures of carbon cores, fluorophores on the shells/surfaces, or the CEE effect but lack definite structures. For a better understanding of the PL center or PL mechanism, further research work on in-depth theoretical investigations and exact structure characterizations including molecular weight, numbers of surface groups, degree of crosslinking, and detailed nature of excited states using advanced techniques (e.g., matrix-assisted laser desorption ionization time-of-flight mass spectroscopy, spherical-aberration correction electron microscopy, synchronous X-ray radiation, and time-resolved electron paramagnetic resonance) are needed to reveal the structure–performance relationship for the development of CDs.

In addition to synthetic methods and the PL mechanism, future research also should focus on designing structures of CDs according to the requirements of applications. (i) CDs, as sensors, have been applied for quantitation of metal ions, organic molecules, or biomolecules, but most of them do not provide higher sensitivity and selectivity as compared to other reported probes, mainly because of their low affinity and low enhancing/quenching efficiency induced by the analyte of interest. While CDs nanocomposites show improved selectivity in sensing and imaging, tedious preparation and separation processes are needed. Thus, functional CDs with high QY, sensitivity, selectivity, and stability should be developed. (ii) For application in anticounterfeiting and data security, the QY, lifetime, and stability of CDs-based afterglow materials should be further improved. Besides, a combined multitechnology for the protection of confidential papers and design of anticounterfeit labels may be the future trend in CDs security encryption. (iii) Applications of CDs in energy remain in infancy, and some key scientific issues need to be addressed before their further development and practical applications. First, it is necessary to establish the relationship among the electrochemical property, morphology, and nanostructure of CDs. Second, exploring the roles of CDs in the electrochemical field through experiments and theoretical calculations is important. Lastly, investigating the assembly behavior, interaction, and reaction mechanism of CDs with other components for the controllable design of high-performance CDs-based electrode materials or catalysts is needed. These are beneficial to illuminate the electrochemical performance and energy storage mechanism of CDs-based materials. (iv) Considering less light scattering, less damage, and deeper light penetration into tissue, CDs emitting deep red to NIR (650–1700 nm) excited by deep red to NIR light are highly

desirable for their future clinical applications in phototheranostics and smart healthcare devices (e.g., the wearable functional brain imaging system, skin temperature monitoring). Therefore, more studies are still needed to develop two-/multiphoton or deep red/NIR emitting CDs with high QY, narrow FWHM, good biocompatibility, high stability, and excellent water solubility for biomedical applications. Moreover, systematic research on toxicity and metabolic pathways of CDs in animal models are critical for their future clinical applications. Furthermore, the exact molecular mechanism of drug-CPDs should be systematically explored.

In summary, this Outlook demonstrates that CDs as a new type of carbon-based nanomaterial possess unique optical features, excellent biocompatibility, low cost, easy modification, and functionalization, and show important and remarkable potential for a wide spectrum of applications. However, from a perspective of CDs-based materials' real applications, there is still a lot of research work to be done. Some in-depth perspectives regarding preparation, mechanism, structures, properties, and applications of CDs have been presented for possible guidance. With the development of advanced technology and characterizations, we believe that controllable synthetic methods, large-scale production, and a better understanding on the structure–performance relationship can be realized, which will greatly extend the application scope of CDs-based materials. Unique features will bring the bright future of CDs.

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Notes

The authors declare no competing financial interest.

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REFERENCES

- (1) Yan, Y.; Gong, J.; Chen, J.; Zeng, Z.; Huang, W.; Pu, K.; Liu, J.; Chen, P. Recent Advances on Graphene Quantum Dots: From Chemistry and Physics to Applications. *Adv. Mater.* **2019**, *31*, 1808283.
- (2) Liu, J.; Geng, Y.; Li, D.; Yao, H.; Huo, Z.; Li, Y.; Zhang, K.; Zhu, S.; Wei, H.; Xu, W.; Jiang, J.; Yang, B. Deep Red Emissive Carbonized

Polymer Dots with Unprecedented Narrow Full Width at Half Maximum. *Adv. Mater.* **2020**, *32*, 1906641.

- (3) Jiang, K.; Wang, Y.; Gao, X.; Cai, C.; Lin, H. Facile, Quick, and Gram-Scale Synthesis of Ultralong-Lifetime Room-Temperature-Phosphorescent Carbon Dots by Microwave Irradiation. *Angew. Chem., Int. Ed.* **2018**, *57*, 6216–6220.

- (4) Hu, C.; Li, M.; Qiu, J.; Sun, Y. P. Design and Fabrication of Carbon Dots for Energy Conversion and Storage. *Chem. Soc. Rev.* **2019**, *48*, 2315–2337.

- (5) Liu, M. L.; Chen, B. B.; Li, C. M.; Huang, C. Z. Carbon Dots: Synthesis, Formation Mechanism, Fluorescence Origin and Sensing Applications. *Green Chem.* **2019**, *21*, 449–471.

- (6) Zhu, S.; Song, Y.; Zhao, X.; Shao, J.; Zhang, J.; Yang, B. The Photoluminescence Mechanism in Carbon Dots (Graphene Quantum Dots, Carbon Nanodots, and Polymer Dots): Current State and Future Perspective. *Nano Res.* **2015**, *8*, 355–381.

- (7) Hoang, V. C.; Dave, K.; Gomes, V. G. Carbon Quantum Dot-Based Composites for Energy Storage and Electrocatalysis: Mechanism, Applications and Future Prospects. *Nano Energy* **2019**, *66*, 104093.

- (8) Zhang, Z.; Yi, G.; Li, P.; Zhang, X.; Fan, H.; Zhang, Y.; Wang, X.; Zhang, C. A Minireview on Doped Carbon Dots for Photocatalytic and Electrocatalytic Applications. *Nanoscale* **2020**, *12*, 13899–13906.

- (9) Yuan, F.; Wang, Y. K.; Sharma, G.; Dong, Y.; Zheng, X.; Li, P.; Johnston, A.; Bappi, G.; Fan, J. Z.; Kung, H.; Chen, B.; Saidaminov, M. I.; Singh, K.; Voznyy, O.; Bakr, O. M.; Lu, Z. H.; Sargent, E. H. Bright High-Colour-Purity Deep-Blue Carbon Dot Light-Emitting Diodes via Efficient Edge Amination. *Nat. Photonics* **2020**, *14*, 171–176.

- (10) Xu, X.; Ray, R.; Gu, Y.; Ploehn, H. J.; Gearheart, L.; Raker, K.; Scrivens, W. A. Electrophoretic Analysis and Purification of Fluorescent Single-Walled Carbon Nanotube Fragments. *J. Am. Chem. Soc.* **2004**, *126*, 12736–12737.

- (11) Sun, Y. P.; Zhou, B.; Lin, Y.; Wang, W.; Fernando, K. A. S.; Pathak, P.; Mezziani, M. J.; Harruff, B. A.; Wang, X.; Wang, H.; Luo, P. G.; Yang, H.; Kose, M. E.; Chen, B.; Veca, L. M.; Xie, S. Y. Quantum-Sized Carbon Dots for Bright and Colorful Photoluminescence. *J. Am. Chem. Soc.* **2006**, *128*, 7756–7757.

- (12) Zhu, S.; Meng, Q.; Wang, L.; Zhang, J.; Song, Y.; Jin, H.; Zhang, K.; Sun, H.; Wang, H.; Yang, B. Highly Photoluminescent Carbon Dots for Multicolor Patterning, Sensors, and Bioimaging. *Angew. Chem., Int. Ed.* **2013**, *52*, 3953–3957.

- (13) Ding, H.; Yu, S. B.; Wei, J. S.; Xiong, H. M. Full-Color Light-Emitting Carbon Dots with a Surface-State-Controlled Luminescence Mechanism. *ACS Nano* **2016**, *10*, 484–491.

- (14) Miao, X.; Qu, D.; Yang, D.; Nie, B.; Zhao, Y.; Fan, H.; Sun, Z. Synthesis of Carbon Dots with Multiple Color Emission by Controlled Graphitization and Surface Functionalization. *Adv. Mater.* **2018**, *30*, 1704740.

- (15) Li, D.; Jing, P.; Sun, L.; An, Y.; Shan, X.; Lu, X.; Zhou, D.; Han, D.; Shen, D.; Zhai, Y.; Qu, S.; Zbořil, R.; Rogach, A. L. Near-Infrared Excitation/Emission and Multiphoton-Induced Fluorescence of Carbon Dots. *Adv. Mater.* **2018**, *30*, 1705913.

- (16) Lu, S.; Sui, L.; Liu, J.; Zhu, S.; Chen, A.; Jin, M.; Yang, B. Near-Infrared Photoluminescent Polymer–Carbon Nanodots with Two-Photon Fluorescence. *Adv. Mater.* **2017**, *29*, 1603443.

- (17) Shao, J.; Zhu, S.; Liu, H.; Song, Y.; Tao, S.; Yang, B. Full-Color Emission Polymer Carbon Dots with Quench-Resistant Solid-State Fluorescence. *Adv. Sci.* **2017**, *4*, 1700395.

- (18) Bao, X.; Yuan, Y.; Chen, J.; Zhang, B.; Li, D.; Zhou, D.; Jing, P.; Xu, G.; Wang, Y.; Holá, K.; Shen, D.; Wu, C.; Song, L.; Liu, C.; Zbořil, R.; Qu, S. In Vivo Theranostics with Near-Infrared-Emitting Carbon Dots—Highly Efficient Photothermal Therapy Based on Passive Targeting after Intravenous Administration. *Light: Sci. Appl.* **2018**, *7*, 91.

- (19) Liu, J.; Li, D.; Zhang, K.; Yang, M.; Sun, H.; Yang, B. One-Step Hydrothermal Synthesis of Nitrogen-Doped Conjugated Carbonized Polymer Dots with 31% Efficient Red Emission for In Vivo Imaging. *Small* **2018**, *14*, 1703919.

- (20) Yuan, F.; Yuan, T.; Sui, L.; Wang, Z.; Xi, Z.; Li, Y.; Li, X.; Fan, L.; Tan, Z.; Chen, A.; Jin, M.; Yang, S. Engineering Triangular Carbon Quantum Dots with Unprecedented Narrow Bandwidth Emission for Multicolored LEDs. *Nat. Commun.* **2018**, *9*, 2249.
- (21) Ye, X.; Xiang, Y.; Wang, Q.; Li, Z.; Liu, Z. A Red Emissive Two-Photon Fluorescence Probe Based on Carbon Dots for Intracellular pH Detection. *Small* **2019**, *15*, 1901673.
- (22) Zhang, M.; Wang, H.; Wang, B.; Ma, Y.; Huang, H.; Liu, Y.; Shao, M.; Yao, B.; Kang, Z. Maltase Decorated by Chiral Carbon Dots with Inhibited Enzyme Activity for Glucose Level Control. *Small* **2019**, *15*, 1901512.
- (23) Li, F.; Li, Y.; Yang, X.; Han, X.; Jiao, Y.; Wei, T.; Yang, D.; Xu, H.; Nie, G. Highly Fluorescent Chiral N-S-Doped Carbon Dots from Cysteine: Affecting Cellular Energy Metabolism. *Angew. Chem., Int. Ed.* **2018**, *57*, 2377–2382.
- (24) Tao, S.; Lu, S.; Geng, Y.; Zhu, S.; Redfern, S. A. T.; Song, Y.; Feng, T.; Xu, W.; Yang, B. Design of Metal-Free Polymer Carbon Dots: A New Class of Room-Temperature Phosphorescent Materials. *Angew. Chem., Int. Ed.* **2018**, *57*, 2393–2398.
- (25) Li, W.; Zhou, W.; Zhou, Z.; Zhang, H.; Zhang, X.; Zhuang, J.; Liu, Y.; Lei, B.; Hu, C. A Universal Strategy for Activating the Multicolor Room-Temperature Afterglow of Carbon Dots in a Boric Acid Matrix. *Angew. Chem., Int. Ed.* **2019**, *58*, 7278–7283.
- (26) Xia, C.; Tao, S.; Zhu, S.; Song, Y.; Feng, T.; Zeng, Q.; Liu, J.; Yang, B. Hydrothermal Addition Polymerization for Ultrahigh-Yield Carbonized Polymer Dots with Room Temperature Phosphorescence via Nanocomposite. *Chem. - Eur. J.* **2018**, *24*, 11303–11308.
- (27) Jiang, K.; Hu, S.; Wang, Y.; Li, Z.; Lin, H. Photo-Stimulated Polychromatic Room Temperature Phosphorescence of Carbon Dots. *Small* **2020**, *16*, 2001909.
- (28) Park, M.; Kim, H. S.; Yoon, H.; Kim, J.; Lee, S.; Yoo, S.; Jeon, S. Controllable Singlet–Triplet Energy Splitting of Graphene Quantum Dots through Oxidation: From Phosphorescence to TADF. *Adv. Mater.* **2020**, *32*, 2000936.
- (29) Zhu, S.; Song, Y.; Shao, J.; Zhao, X.; Yang, B. Non-Conjugated Polymer Dots with Crosslink-Enhanced Emission in the Absence of Fluorophore Units. *Angew. Chem., Int. Ed.* **2015**, *54*, 14626–14637.
- (30) Pan, D.; Zhang, J.; Li, Z.; Wu, M. Hydrothermal Route for Cutting Graphene Sheets into Blue-Luminescent Graphene Quantum Dots. *Adv. Mater.* **2010**, *22*, 734–738.
- (31) Zhu, S.; Zhang, J.; Tang, S.; Qiao, C.; Wang, L.; Wang, H.; Liu, X.; Li, B.; Li, Y.; Yu, W.; Wang, X.; Sun, H.; Yang, B. Surface Chemistry Routes to Modulate the Photoluminescence of Graphene Quantum Dots: From Fluorescence Mechanism to up-Conversion Bioimaging Applications. *Adv. Funct. Mater.* **2012**, *22*, 4732–4740.
- (32) Peng, J.; Gao, W.; Gupta, B. K.; Liu, Z.; Romero-Aburto, R.; Ge, L.; Song, L.; Alemany, L. B.; Zhan, X.; Gao, G.; Vithayathil, S. A.; Kaiparettu, B. A.; Marti, A. A.; Hayashi, T.; Zhu, J. J.; Ajayan, P. M. Graphene Quantum Dots Derived from Carbon Fibers. *Nano Lett.* **2012**, *12*, 844–849.
- (33) Li, Y.; Hu, Y.; Zhao, Y.; Shi, G.; Deng, L.; Hou, Y.; Qu, L. An Electrochemical Avenue to Green-Luminescent Graphene Quantum Dots as Potential Electron-Acceptors for Photovoltaics. *Adv. Mater.* **2011**, *23*, 776–780.
- (34) Li, L.; Wu, G.; Yang, G.; Peng, J.; Zhao, J.; Zhu, J. J. Focusing on Luminescent Graphene Quantum Dots: Current Status and Future Perspectives. *Nanoscale* **2013**, *5*, 4015–4039.
- (35) Zhu, S.; Zhang, J.; Qiao, C.; Tang, S.; Li, Y.; Yuan, W.; Li, B.; Tian, L.; Liu, F.; Hu, R.; Gao, H.; Wei, H.; Zhang, H.; Sun, H.; Yang, B. Strongly Green-Photoluminescent Graphene Quantum Dots for Bioimaging Applications. *Chem. Commun.* **2011**, *47*, 6858–6860.
- (36) Song, Y.; Zhu, S.; Zhang, S.; Fu, Y.; Wang, L.; Zhao, X.; Yang, B. Investigation from Chemical Structure to Photoluminescent Mechanism: A Type of Carbon Dots from the Pyrolysis of Citric Acid and an Amine. *J. Mater. Chem. C* **2015**, *3*, 5976–5984.
- (37) Xia, C.; Zhu, S.; Feng, T.; Yang, M.; Yang, B. Evolution and Synthesis of Carbon Dots: From Carbon Dots to Carbonized Polymer Dots. *Adv. Sci.* **2019**, *6*, 1901316.
- (38) Feng, T.; Zeng, Q.; Lu, S.; Yan, X.; Liu, J.; Tao, S.; Yang, M.; Yang, B. Color-Tunable Carbon Dots Possessing Solid-State Emission for Full-Color Light-Emitting Diodes Applications. *ACS Photonics* **2018**, *5*, 502–510.
- (39) Feng, T.; Zhu, S.; Zeng, Q.; Lu, S.; Tao, S.; Liu, J.; Yang, B. Supramolecular Cross-Link-Regulated Emission and Related Applications in Polymer Carbon Dots. *ACS Appl. Mater. Interfaces* **2018**, *10*, 12262–12277.
- (40) Han, M.; Zhu, S.; Lu, S.; Song, Y.; Feng, T.; Tao, S.; Liu, J.; Yang, B. Recent Progress on the Photocatalysis of Carbon Dots: Classification, Mechanism and Applications. *Nano Today* **2018**, *19*, 201–218.
- (41) Chen, Y.; Cao, Y.; Ma, C.; Zhu, J. J. Carbon-Based Dots for Electrochemiluminescence Sensing. *Mater. Chem. Front.* **2020**, *4*, 369–385.
- (42) Jiang, K.; Wang, Y.; Li, Z.; Lin, H. Afterglow of Carbon Dots: Mechanism, Strategy and Applications. *Mater. Chem. Front.* **2020**, *4*, 386–399.
- (43) Sui, B.; Li, Y.; Yang, B. Nanocomposite Hydrogels Based on Carbon Dots and Polymers. *Chinese. Chin. Chem. Lett.* **2020**, *31*, 1443–1447.
- (44) Jiang, F.; Chen, D.; Li, R.; Wang, Y.; Zhang, G.; Li, S.; Zheng, J.; Huang, N.; Gu, Y.; Wang, C.; Shu, C. Eco-Friendly Synthesis of Size-Controllable Amine-Functionalized Graphene Quantum Dots with Antimycoplasmal Properties. *Nanoscale* **2013**, *5*, 1137–1142.
- (45) Liu, J. H.; Li, R. S.; Yuan, B.; Wang, J.; Li, Y. F.; Huang, C. Z. Mitochondria-Targeting Single-Layered Graphene Quantum Dots with Dual Recognition Sites for ATP Imaging in Living Cells. *Nanoscale* **2018**, *10*, 17402–17408.
- (46) Xu, X.; Tang, W.; Zhou, Y.; Bao, Z.; Su, Y.; Hu, J.; Zeng, H. Steering Photoelectrons Excited in Carbon Dots into Platinum Cluster Catalyst for Solar-Driven Hydrogen Production. *Adv. Sci.* **2017**, *4*, 1700273.
- (47) Huang, X.; Zhang, F.; Zhu, L.; Choi, K. Y.; Guo, N.; Guo, J.; Tackett, K.; Anilkumar, P.; Liu, G.; Quan, Q.; Choi, H. S.; Niu, G.; Sun, Y. P.; Lee, S.; Chen, X. Effect of Injection Routes on the Biodistribution, Clearance, and Tumor Uptake of Carbon Dots. *ACS Nano* **2013**, *7*, 5684–5693.
- (48) Yang, M.; Tang, Q.; Meng, Y.; Liu, J.; Feng, T.; Zhao, X.; Zhu, S.; Yu, W.; Yang, B. Reversible “off-On” Fluorescence of Zn²⁺-Passivated Carbon Dots: Mechanism and Potential for the Detection of EDTA and Zn²⁺. *Langmuir* **2018**, *34*, 7767–7775.
- (49) Yuan, F.; Wang, Z.; Li, X.; Li, Y.; Tan, Z.; Fan, L.; Yang, S. Bright Multicolor Bandgap Fluorescent Carbon Quantum Dots for Electroluminescent Light-Emitting Diodes. *Adv. Mater.* **2017**, *29*, 1604436.
- (50) Qu, S.; Zhou, D.; Li, D.; Ji, W.; Jing, P.; Han, D.; Liu, L.; Zeng, H.; Shen, D. Toward Efficient Orange Emissive Carbon Nanodots through Conjugated Sp²-Domain Controlling and Surface Charges Engineering. *Adv. Mater.* **2016**, *28*, 3516–3521.
- (51) Gao, T.; Wang, X.; Yang, L. Y.; He, H.; Ba, X. X.; Zhao, J.; Jiang, F. L.; Liu, Y. Red, Yellow, and Blue Luminescence by Graphene Quantum Dots: Syntheses, Mechanism, and Cellular Imaging. *ACS Appl. Mater. Interfaces* **2017**, *9*, 24846–24856.
- (52) Sun, S.; Zhang, L.; Jiang, K.; Wu, A.; Lin, H. Toward High-Efficient Red Emissive Carbon Dots: Facile Preparation, Unique Properties, and Applications as Multifunctional Theranostic Agents. *Chem. Mater.* **2016**, *28*, 8659–8668.
- (53) Fang, B. Y.; Li, C.; Song, Y. Y.; Tan, F.; Cao, Y. C.; Zhao, Y. Di. Nitrogen-Doped Graphene Quantum Dot for Direct Fluorescence Detection of Al³⁺ in Aqueous Media and Living Cells. *Biosens. Bioelectron.* **2018**, *100*, 41–48.
- (54) Tetsuka, H.; Asahi, R.; Nagoya, A.; Okamoto, K.; Tajima, I.; Ohta, R.; Okamoto, A. Optically Tunable Amino-Functionalized Graphene Quantum Dots. *Adv. Mater.* **2012**, *24*, 5333–5338.
- (55) Kang, Z.; Lee, S. Carbon Dots: Advances in Nanocarbon Applications. *Nanoscale* **2019**, *11*, 19214–19224.
- (56) Zhang, Y.; Liu, X.; Fan, Y.; Guo, X.; Zhou, L.; Lv, Y.; Lin, J. One-Step Microwave Synthesis of N-Doped Hydroxyl-Functionalized

Carbon Dots with Ultra-High Fluorescence Quantum Yields. *Nanoscale* **2016**, *8*, 15281–15287.

(57) Hui, W.; Yang, Y.; Xu, Q.; Gu, H.; Feng, S.; Su, Z.; Zhang, M.; Wang, J.; Li, X.; Fang, J.; Xia, F.; Xia, Y.; Chen, Y.; Gao, X.; Huang, W. Red-Carbon-Quantum-Dot-Doped SnO₂ Composite with Enhanced Electron Mobility for Efficient and Stable Perovskite Solar Cells. *Adv. Mater.* **2020**, *32*, 1906374.

(58) Zheng, M.; Li, Y.; Liu, S.; Wang, W.; Xie, Z.; Jing, X. One-Pot to Synthesize Multifunctional Carbon Dots for Near Infrared Fluorescence Imaging and Photothermal Cancer Therapy. *ACS Appl. Mater. Interfaces* **2016**, *8*, 23533–23541.

(59) Yang, M.; Meng, Y.; Liu, J.; Yu, W.; Yang, B. Facile Synthesis of Mg²⁺-Doped Carbon Dots as Novel Biomaterial Inducing Cell Osteoblastic Differentiation. *Part. Part. Syst. Charact.* **2019**, *36*, 1800315.

(60) Liu, J.; Lu, S.; Tang, Q.; Zhang, K.; Yu, W.; Sun, H.; Yang, B. One-Step Hydrothermal Synthesis of Photoluminescent Carbon Nanodots with Selective Antibacterial Activity against *Porphyromonas gingivalis*. *Nanoscale* **2017**, *9*, 7135–7142.

(61) Ke, C.-B.; Lu, T.-L.; Chen, J.-L. Fluorometric Determination of Amifostine and Alkaline Phosphatase on Amphiprotic Molecularly Imprinted Silica Crosslinked with Binary Functional Silanes and Carbon Dots. *Biosens. Bioelectron.* **2020**, *151*, 111965.

(62) Han, Z.; Ni, Y.; Ren, J.; Zhang, W.; Wang, Y.; Xie, Z.; Zhou, S.; Yu, S. F. Orange Emissive Carbon Dots for Microcavity. *Nanoscale* **2019**, *11*, 11577–11583.

(63) Gan, N.; Shi, H.; An, Z.; Huang, W. Recent Advances in Polymer-Based Metal-Free Room-Temperature Phosphorescent Materials. *Adv. Funct. Mater.* **2018**, *28*, 1802657.

(64) Su, Q.; Lu, C.; Yang, X. Efficient Room Temperature Phosphorescence Carbon Dots: Information Encryption and Dual-Channel PH Sensing. *Carbon* **2019**, *152*, 609–615.

(65) Jiang, K.; Zhang, L.; Lu, J.; Xu, C.; Cai, C.; Lin, H. Luminescent Dyes Very Important Paper Triple-Mode Emission of Carbon Dots: Applications for Advanced. *Angew. Chem., Int. Ed.* **2016**, *55*, 7231–7235.

(66) Zhang, D. Y.; Zheng, Y.; Zhang, H.; He, L.; Tan, C. P.; Sun, J. H.; Zhang, W.; Peng, X.; Zhan, Q.; Ji, L. N.; Mao, Z. W. Ruthenium Complex-Modified Carbon Nanodots for Lysosome-Targeted One- and Two-Photon Imaging and Photodynamic Therapy. *Nanoscale* **2017**, *9*, 18966–18976.

(67) Lan, M.; Zhao, S.; Zhang, Z.; Yan, L.; Guo, L.; Niu, G.; Zhang, J.; Zhao, J.; Zhang, H.; Wang, P.; Zhu, G.; Lee, C. S.; Zhang, W. Two-Photon-Excited near-Infrared Emissive Carbon Dots as Multifunctional Agents for Fluorescence Imaging and Photothermal Therapy. *Nano Res.* **2017**, *10*, 3113–3123.

(68) Jie, G.; Zhou, Q.; Jie, G. Graphene Quantum Dots-Based Electrochemiluminescence Detection of DNA Using Multiple Cycling Amplification Strategy. *Talanta* **2019**, *194*, 658–663.

(69) Molaei, M. J. A Review on Nanostructured Carbon Quantum Dots and Their Applications in Biotechnology, Sensors, and Chemiluminescence. *Talanta* **2019**, *196*, 456–478.

(70) Shen, C. L.; Lou, Q.; Zang, J. H.; Liu, K. K.; Qu, S. N.; Dong, L.; Shan, C. X. Near-Infrared Chemiluminescent Carbon Nanodots and Their Application in Reactive Oxygen Species Bioimaging. *Adv. Sci.* **2020**, *7*, 1903525.

(71) Dhenadhayan, N.; Lin, K. C.; Saleh, T. A. Recent Advances in Functionalized Carbon Dots toward the Design of Efficient Materials for Sensing and Catalysis Applications. *Small* **2020**, *16*, 1905767.

(72) Gao, W.; Song, H.; Wang, X.; Liu, X.; Pang, X.; Zhou, Y.; Gao, B.; Peng, X. Carbon Dots with Red Emission for Sensing of Pt²⁺, Au³⁺, and Pd²⁺ and Their Bioapplications in Vitro and in Vivo. *ACS Appl. Mater. Interfaces* **2018**, *10*, 1147–1154.

(73) Zhang, M.; Wang, W.; Yuan, P.; Chi, C.; Zhang, J.; Zhou, N. Synthesis of Lanthanum Doped Carbon Dots for Detection of Mercury Ion, Multi-Color Imaging of Cells and Tissue, and Bacteriostasis. *Chem. Eng. J.* **2017**, *330*, 1137–1147.

(74) Lesani, P.; Singh, G.; Viray, C. M.; Ramaswamy, Y.; Zhu, D. M.; Kingshott, P.; Lu, Z.; Zreiqat, H. Two-Photon Dual-Emissive Carbon

Dot-Based Probe: Deep-Tissue Imaging and Ultrasensitive Sensing of Intracellular Ferric Ions. *ACS Appl. Mater. Interfaces* **2020**, *12*, 18395–18406.

(75) Liu, J.; Dong, Y.; Ma, Y.; Han, Y.; Ma, S.; Chen, H.; Chen, X. One-Step Synthesis of Red/Green Dual-Emissive Carbon Dots for Ratiometric Sensitive ONOO⁻ Probing and Cell Imaging. *Nanoscale* **2018**, *10*, 13589–13598.

(76) Wei, Z.; Li, H.; Liu, S.; Wang, W.; Chen, H.; Xiao, L.; Ren, C.; Chen, X. Carbon Dots as Fluorescent/Colorimetric Probes for Real-Time Detection of Hypochlorite and Ascorbic Acid in Cells and Body Fluid. *Anal. Chem.* **2019**, *91*, 15477–15483.

(77) Yang, P.; Zhu, Z.; Zhang, T.; Zhang, W.; Chen, W.; Cao, Y.; Chen, M.; Zhou, X. Orange-Emissive Carbon Quantum Dots: Toward Application in Wound PH Monitoring Based on Colorimetric and Fluorescent Changing. *Small* **2019**, *15*, 1902823.

(78) Gao, Y.; Jiao, Y.; Lu, W.; Liu, Y.; Han, H.; Gong, X.; Xian, M.; Shuang, S.; Dong, C. Carbon Dots with Red Emission as a Fluorescent and Colorimetric Dual-Readout Probe for the Detection of Chromium(vi) and Cysteine and Its Logic Gate Operation. *J. Mater. Chem. B* **2018**, *6*, 6099–6107.

(79) Jiao, Y.; Gao, Y.; Meng, Y.; Lu, W.; Liu, Y.; Han, H.; Shuang, S.; Li, L.; Dong, C. One-Step Synthesis of Label-Free Ratiometric Fluorescence Carbon Dots for the Detection of Silver Ions and Glutathione and Cellular Imaging Applications. *ACS Appl. Mater. Interfaces* **2019**, *11*, 16822–16829.

(80) Yao, W.; Wu, N.; Lin, Z.; Chen, J.; Li, S.; Weng, S.; Zhang, L.; Liu, A.; Lin, X. Fluorescent Turn-off Competitive Immunoassay for Biotin Based on Hydrothermally Synthesized Carbon Dots. *Microchim. Acta* **2017**, *184*, 907–914.

(81) Sun, Z.; Chen, Z.; Luo, J.; Zhu, Z.; Zhang, X.; Liu, R.; Wu, Z.-c. A Yellow-Emitting Nitrogen-Doped Carbon Dots for Sensing of Vitamin B12 and Their Cell-Imaging. *Dyes Pigm.* **2020**, *176*, 108227.

(82) Hu, G.; Ge, L.; Li, Y.; Mukhtar, M.; Shen, B.; Yang, D.; Li, J. Carbon Dots Derived from Flax Straw for Highly Sensitive and Selective Detections of Cobalt, Chromium, and Ascorbic Acid. *J. Colloid Interface Sci.* **2020**, *579*, 96–108.

(83) Liu, H.; Sun, Y.; Li, Z.; Yang, J.; Aryee, A. A.; Qu, L.; Du, D.; Lin, Y. Lysosome-Targeted Carbon Dots for Ratiometric Imaging of Formaldehyde in Living Cells. *Nanoscale* **2019**, *11*, 8458–8463.

(84) Gu, J.; Li, X.; Zhou, Z.; Liu, W.; Li, K.; Gao, J.; Zhao, Y.; Wang, Q. 2D MnO₂ Nanosheets Generated Signal Transduction with 0D Carbon Quantum Dots: Synthesis Strategy, Dual-Mode Behavior and Glucose Detection. *Nanoscale* **2019**, *11*, 13058–13068.

(85) Gong, P.; Sun, L.; Wang, F.; Liu, X.; Yan, Z.; Wang, M.; Zhang, L.; Tian, Z.; Liu, Z.; You, J. Highly Fluorescent N-Doped Carbon Dots with Two-Photon Emission for Ultrasensitive Detection of Tumor Marker and Visual Monitor Anticancer Drug Loading and Delivery. *Chem. Eng. J.* **2019**, *356*, 994–1002.

(86) Cui, Y.; Liu, R.; Ye, F.; Zhao, S. Single-Excitation, Dual-Emission Biomass Quantum Dots: Preparation and Application for Ratiometric Fluorescence Imaging of Coenzyme A in Living Cells. *Nanoscale* **2019**, *11*, 9270–9275.

(87) Yang, D.; Guo, Z.; Wang, J.; Jin, Y.; Mei, Q.; Miao, P. Carbon Nanodot-Based Fluorescent Method for Virus DNA Analysis with Isothermal Strand Displacement Amplification. *Part. Part. Syst. Charact.* **2019**, *36*, 1900273.

(88) Gao, G.; Jiang, Y. W.; Yang, J.; Wu, F. G. Mitochondria-Targetable Carbon Quantum Dots for Differentiating Cancerous Cells from Normal Cells. *Nanoscale* **2017**, *9*, 18368–18378.

(89) Cheng, W.; Xu, J.; Guo, Z.; Yang, D.; Chen, X.; Yan, W.; Miao, P. Hydrothermal Synthesis of N,S Co-Doped Carbon Nanodots for Highly Selective Detection of Living Cancer Cells. *J. Mater. Chem. B* **2018**, *6*, 5775–5780.

(90) Zhao, X.; Tang, Q.; Zhu, S.; Bu, W.; Yang, M.; Liu, X.; Meng, Y.; Yu, W.; Sun, H.; Yang, B. Controllable Acidophilic Dual-Emission Fluorescent Carbonized Polymer Dots for Selective Imaging of Bacteria. *Nanoscale* **2019**, *11*, 9526–9532.

- (91) Cui, Y.; Hegde, R. S.; Phang, I. Y.; Lee, H. K.; Ling, X. Y. Encoding Molecular Information in Plasmonic Nanostructures for Anti-Counterfeiting Applications. *Nanoscale* **2014**, *6*, 282–288.
- (92) Wang, B.; Yu, Y.; Zhang, H.; Xuan, Y.; Chen, G.; Ma, W.; Li, J.; Yu, J. Carbon Dots in a Matrix: Energy-Transfer-Enhanced Room-Temperature Red Phosphorescence. *Angew. Chem., Int. Ed.* **2019**, *58*, 18443–18448.
- (93) Yang, M.; Feng, T.; Chen, Y.; Liu, J.; Zhao, X.; Yang, B. Synchronously Integration of Co, Fe Dual-Metal Doping in Ru@C and CDs for Boosted Water Splitting Performances in Alkaline Media. *Appl. Catal., B* **2020**, *267*, 118657.
- (94) Cailotto, S.; Negrato, M.; Daniele, S.; Luque, R.; Selva, M.; Amadio, E.; Perosa, A. Carbon Dots as Photocatalysts for Organic Synthesis: Metal-Free Methylene-Oxygen-Bond Photocleavage. *Green Chem.* **2020**, *22*, 1145–1149.
- (95) Han, M.; Lu, S.; Qi, F.; Zhu, S.; Sun, H.; Yang, B. Carbon Dots–Implanted Graphitic Carbon Nitride Nanosheets for Photocatalysis: Simultaneously Manipulating Carrier Transport in Inter- and Intralayers. *Sol. RRL* **2020**, *4*, 1900517.
- (96) Xu, L.; Bai, X.; Guo, L.; Yang, S.; Jin, P.; Yang, L. Facial Fabrication of Carbon Quantum Dots (CDs)-Modified N-TiO₂-x Nanocomposite for the Efficient Photoreduction of Cr(VI) under Visible Light. *Chem. Eng. J.* **2019**, *357*, 473–486.
- (97) Wang, S.; Li, L.; Zhu, Z.; Zhao, M.; Zhang, L.; Zhang, N.; Wu, Q.; Wang, X.; Li, G. Remarkable Improvement in Photocatalytic Performance for Tannery Wastewater Processing via SnS₂ Modified with N-Doped Carbon Quantum Dots: Synthesis, Characterization, and 4-Nitrophenol-Aided Cr(VI) Photoreduction. *Small* **2019**, *15*, 1804515.
- (98) Huang, S.; Jiang, S.; Pang, H.; Wen, T.; Asiri, A. M.; Alamry, K. A.; Alsaedi, A.; Wang, X.; Wang, S. Dual Functional Nanocomposites of Magnetic MnFe₂O₄ and Fluorescent Carbon Dots for Efficient U(VI) Removal. *Chem. Eng. J.* **2019**, *368*, 941–950.
- (99) Luo, H.; Liu, Y.; Dimitrov, S. D.; Steier, L.; Guo, S.; Li, X.; Feng, J.; Xie, F.; Fang, Y.; Sapelkin, A.; Wang, X.; Titirici, M.-M. Pt Single-Atoms Supported on Nitrogen-Doped Carbon Dots for Highly Efficient Photocatalytic Hydrogen Generation. *J. Mater. Chem. A* **2020**, *8*, 14690–14696.
- (100) Yan, Y.; Chen, J.; Li, N.; Tian, J.; Li, K.; Jiang, J.; Liu, J.; Tian, Q.; Chen, P. Systematic Bandgap Engineering of Graphene Quantum Dots and Applications for Photocatalytic Water Splitting and CO₂ Reduction. *ACS Nano* **2018**, *12*, 3523–3532.
- (101) Li, M.; Wang, M.; Zhu, L.; Li, Y.; Yan, Z.; Shen, Z.; Cao, X. Facile Microwave Assisted Synthesis of N-Rich Carbon Quantum Dots/Dual-Phase TiO₂ Heterostructured Nanocomposites with High Activity in CO₂ Photoreduction. *Appl. Catal., B* **2018**, *231*, 269–276.
- (102) Sarma, D.; Majumdar, B.; Sarma, T. K. Visible-Light Induced Enhancement in the Multi-Catalytic Activity of Sulfated Carbon Dots for Aerobic Carbon-Carbon Bond Formation. *Green Chem.* **2019**, *21*, 6717–6726.
- (103) Bhattacharyya, S.; Ehrat, F.; Urban, P.; Teves, R.; Wyrwich, R.; Döblinger, M.; Feldmann, J.; Urban, A. S.; Stolarczyk, J. K. Effect of Nitrogen Atom Positioning on the Trade-off between Emissive and Photocatalytic Properties of Carbon Dots. *Nat. Commun.* **2017**, *8*, 1401.
- (104) Guo, S.; Zhao, S.; Wu, X.; Li, H.; Zhou, Y.; Zhu, C.; Yang, N.; Jiang, X.; Gao, J.; Bai, L.; Liu, Y.; Lifshitz, Y.; Lee, S. T.; Kang, Z. A Co₃O₄-CDots-C₃N₄ Three Component Electrocatalyst Design Concept for Efficient and Tunable CO₂ Reduction to Syngas. *Nat. Commun.* **2017**, *8*, 1828.
- (105) Liu, C.; Fu, Y.; Xia, Y.; Zhu, C.; Hu, L.; Zhang, K.; Wu, H.; Huang, H.; Liu, Y.; Xie, T.; Zhong, J.; Kang, Z. Cascaded Photopotential in a Carbon Dot-Hematite System Driving Overall Water Splitting under Visible Light. *Nanoscale* **2018**, *10*, 2454–2460.
- (106) Han, Y.; Tang, D.; Yang, Y.; Li, C.; Kong, W.; Huang, H.; Liu, Y.; Kang, Z. Non-Metal Single/Dual Doped Carbon Quantum Dots: A General Flame Synthetic Method and Electro-Catalytic Properties. *Nanoscale* **2015**, *7*, 5955–5962.
- (107) Fan, T.; Zhang, G.; Jian, L.; Murtaza, I.; Meng, H.; Liu, Y.; Min, Y. Facile Synthesis of Defect-Rich Nitrogen and Sulfur Co-Doped Graphene Quantum Dots as Metal-Free Electrocatalyst for the Oxygen Reduction Reaction. *J. Alloys Compd.* **2019**, *792*, 844–850.
- (108) Shin, J.; Guo, J.; Zhao, T.; Guo, Z. Functionalized Carbon Dots on Graphene as Outstanding Non-Metal Bifunctional Oxygen Electrocatalyst. *Small* **2019**, *15*, 1900296.
- (109) Tian, J.; Chen, J.; Liu, J.; Tian, Q.; Chen, P. Graphene Quantum Dot Engineered Nickel-Cobalt Phosphide as Highly Efficient Bifunctional Catalyst for Overall Water Splitting. *Nano Energy* **2018**, *48*, 284–291.
- (110) Yang, S.; Du, R.; Yu, Y.; Zhang, Z.; Wang, F. One-Step Electrodeposition of Carbon Quantum Dots and Transition Metal Ions for N-Doped Carbon Coupled with NiFe Oxide Clusters: A High-Performance Electrocatalyst for Oxygen Evolution. *Nano Energy* **2020**, *77*, 105057.
- (111) Raza Naqvi, S. T.; Rasheed, T.; Majeed, S.; Hussain, D.; Fatima, B.; Najam ul Haq, M.; Nawaz, R.; Ahmad, N.; Noon, T. Nitrogen Doped Carbon Quantum Dots Conjugated with AgNi Alloy Nanoparticles as Potential Electrocatalyst for Efficient Water Splitting. *J. Alloys Compd.* **2020**, *847*, 156492.
- (112) Chen, Z.; Mou, K.; Wang, X.; Liu, L. Nitrogen-Doped Graphene Quantum Dots Enhance the Activity of Bi₂O₃ Nanosheets for Electrochemical Reduction of CO₂ in a Wide Negative Potential Region. *Angew. Chem., Int. Ed.* **2018**, *57*, 12790–12794.
- (113) Lv, K.; Suo, W.; Shao, M.; Zhu, Y.; Wang, X.; Feng, J.; Fang, M. Nitrogen Doped MoS₂ and Nitrogen Doped Carbon Dots Composite Catalyst for Electroreduction CO₂ to CO with High Faradaic Efficiency. *Nano Energy* **2019**, *63*, 103834.
- (114) Zhou, T.; Chen, S.; Li, L.; Wang, J.; Zhang, Y.; Li, J.; Bai, J.; Xia, L.; Xu, Q.; Rahim, M.; Zhou, B. Carbon Quantum Dots Modified Anatase/Rutile TiO₂ Photoanode with Dramatically Enhanced Photoelectrochemical Performance. *Appl. Catal., B* **2020**, *269*, 118776.
- (115) Niu, Q.; Gu, X.; Li, L.; Zhang, Y.-n.; Zhao, G. 3D CQDs-{001}TiO₂/Ti Photoelectrode with Dominant {001} Facets: Efficient Visible-Light-Driven Photoelectrocatalytic Oxidation of Organic Pollutants and Mechanism Insight. *Appl. Catal., B* **2020**, *261*, 118229.
- (116) Wu, Y.; Zhang, H.; Pan, A.; Wang, Q.; Zhang, Y.; Zhou, G.; He, L. White-Light-Emitting Melamine-Formaldehyde Microspheres through Polymer-Mediated Aggregation and Encapsulation of Graphene Quantum Dots. *Adv. Sci.* **2019**, *6*, 1801432.
- (117) Wang, C.; Hu, T.; Chen, Y.; Xu, Y.; Song, Q. Polymer-Assisted Self-Assembly of Multicolor Carbon Dots as Solid-State Phosphors for Fabrication of Warm, High-Quality, and Temperature-Responsive White-Light-Emitting Devices. *ACS Appl. Mater. Interfaces* **2019**, *11*, 22332–22338.
- (118) Yan, F.; Jiang, Y.; Sun, X.; Wei, J.; Chen, L.; Zhang, Y. Multicolor Carbon Dots with Concentration-Tunable Fluorescence and Solvent-Affected Aggregation States for White Light-Emitting Diodes. *Nano Res.* **2020**, *13*, 52–60.
- (119) Zhan, Y.; Shang, B.; Chen, M.; Wu, L. One-Step Synthesis of Silica-Coated Carbon Dots with Controllable Solid-State Fluorescence for White Light-Emitting Diodes. *Small* **2019**, *15*, 1901161.
- (120) Zhou, D.; Zhai, Y.; Qu, S.; Li, D.; Jing, P.; Ji, W.; Shen, D.; Rogach, A. L. Electrostatic Assembly Guided Synthesis of Highly Luminescent Carbon-Nanodots@BaSO₄ Hybrid Phosphors with Improved Stability. *Small* **2017**, *13*, 1602055.
- (121) Liu, E.; Li, D.; Zhou, X.; Zhou, G.; Xiao, H.; Zhou, D.; Tian, P.; Guo, R.; Qu, S. Highly Emissive Carbon Dots in Solid State and Their Applications in Light-Emitting Devices and Visible Light Communication. *ACS Sustain. ACS Sustainable Chem. Eng.* **2019**, *7*, 9301–9308.
- (122) Zhou, D.; Jing, P.; Wang, Y.; Zhai, Y.; Li, D.; Xiong, Y.; Baranov, A. V.; Qu, S.; Rogach, A. L. Carbon Dots Produced: Via Space-Confined Vacuum Heating: Maintaining Efficient Luminescence in Both Dispersed and Aggregated States. *Nanoscale Horizons* **2019**, *4*, 388–395.

- (123) He, J.; He, Y.; Chen, Y.; Lei, B.; Zhuang, J.; Xiao, Y.; Liang, Y.; Zheng, M.; Zhang, H.; Liu, Y. Solid-State Carbon Dots with Red Fluorescence and Efficient Construction of Dual-Fluorescence Morphologies. *Small* **2017**, *13*, 1700075.
- (124) Wang, F.; Chen, Y. H.; Liu, C. Y.; Ma, D. G. White Light-Emitting Devices Based on Carbon Dots' Electroluminescence. *Chem. Commun.* **2011**, *47*, 3502–3504.
- (125) Zhou, S.; Tang, R.; Yin, L. Slow-Photon-Effect-Induced Photoelectrical-Conversion Efficiency Enhancement for Carbon-Quantum-Dot-Sensitized Inorganic CsPbBr₃ Inverse Opal Perovskite Solar Cells. *Adv. Mater.* **2017**, *29*, 1703682.
- (126) Bian, H.; Wang, Q.; Yang, S.; Yan, C.; Wang, H.; Liang, L.; Jin, Z.; Wang, G.; Liu, S. Nitrogen-Doped Graphene Quantum Dots for 80% Photoluminescence Quantum Yield for Inorganic γ -CsPbI₃ Perovskite Solar Cells with Efficiency beyond 16%. *J. Mater. Chem. A* **2019**, *7*, 5740–5747.
- (127) Diao, S.; Zhang, X.; Shao, Z.; Ding, K.; Jie, J.; Zhang, X. 12.35% Efficient Graphene Quantum Dots/Silicon Heterojunction Solar Cells Using Graphene Transparent Electrode. *Nano Energy* **2017**, *31*, 359–366.
- (128) Li, Z.; Liu, X.; Wang, L.; Bu, F.; Wei, J.; Pan, D.; Wu, M. Hierarchical 3D All-Carbon Composite Structure Modified with N-Doped Graphene Quantum Dots for High-Performance Flexible Supercapacitors. *Small* **2018**, *14*, 1801498.
- (129) Qing, Y.; Jiang, Y.; Lin, H.; Wang, L.; Liu, A.; Cao, Y.; Sheng, R.; Guo, Y.; Fan, C.; Zhang, S.; Jia, D.; Fan, Z. Boosting the Supercapacitor Performance of Activated Carbon by Constructing Overall Conductive Networks Using Graphene Quantum Dots. *J. Mater. Chem. A* **2019**, *7*, 6021–6027.
- (130) Devadas, B.; Imae, T. Effect of Carbon Dots on Conducting Polymers for Energy Storage Applications. *ACS Sustain. ACS Sustainable Chem. Eng.* **2018**, *6*, 127–134.
- (131) Wei, J. S.; Ding, C.; Zhang, P.; Ding, H.; Niu, X. Q.; Ma, Y. Y.; Li, C.; Wang, Y. G.; Xiong, H. M. Robust Negative Electrode Materials Derived from Carbon Dots and Porous Hydrogels for High-Performance Hybrid Supercapacitors. *Adv. Mater.* **2018**, *31*, 1806197.
- (132) Jia, H.; Cai, Y.; Lin, J.; Liang, H.; Qi, J.; Cao, J.; Feng, J.; Fei, W. D. Heterostructural Graphene Quantum Dot/MnO₂ Nanosheets toward High-Potential Window Electrodes for High-Performance Supercapacitors. *Adv. Sci.* **2018**, *5*, 1700887.
- (133) Strauss, V.; Marsh, K.; Kowal, M. D.; El-Kady, M.; Kaner, R. B. A Simple Route to Porous Graphene from Carbon Nanodots for Supercapacitor Applications. *Adv. Mater.* **2018**, *30*, 1704449.
- (134) Zhang, Y.; Foster, C. W.; Banks, C. E.; Shao, L.; Hou, H.; Zou, G.; Chen, J.; Huang, Z.; Ji, X. Graphene-Rich Wrapped Petal-Like Rutile TiO₂ Tuned by Carbon Dots for High-Performance Sodium Storage. *Adv. Mater.* **2016**, *28*, 9391–9399.
- (135) Zhang, E.; Jia, X.; Wang, B.; Wang, J.; Yu, X.; Lu, B. Carbon Dots@rGO Paper as Freestanding and Flexible Potassium-Ion Batteries Anode. *Adv. Sci.* **2020**, *7*, 2000470.
- (136) Zhang, Q.; Sun, C.; Fan, L.; Zhang, N.; Sun, K. Iron Fluoride Vertical Nanosheets Array Modified with Graphene Quantum Dots as Long-Life Cathode for Lithium Ion Batteries. *Chem. Eng. J.* **2019**, *371*, 245–251.
- (137) Yin, X.; Chen, H.; Zhi, C.; Sun, W.; Lv, L. P.; Wang, Y. Functionalized Graphene Quantum Dot Modification of Yolk–Shell NiO Microspheres for Superior Lithium Storage. *Small* **2018**, *14*, 1800589.
- (138) Ma, C.; Dai, K.; Hou, H.; Ji, X.; Chen, L.; Ivey, D. G.; Wei, W. High Ion-Conducting Solid-State Composite Electrolytes with Carbon Quantum Dot Nanofillers. *Adv. Sci.* **2018**, *5*, 1700996.
- (139) Zheng, C.; Luo, N.; Huang, S.; Wu, W.; Huang, H.; Wei, M. Nanocomposite of Mo₂N Quantum Dots@MoO₃@Nitrogen-Doped Carbon as a High-Performance Anode for Lithium-Ion Batteries. *ACS Sustain. ACS Sustainable Chem. Eng.* **2019**, *7*, 10198–10206.
- (140) Hu, Y.; Chen, W.; Lei, T.; Zhou, B.; Jiao, Y.; Yan, Y.; Du, X.; Huang, J.; Wu, C.; Wang, X.; Wang, Y.; Chen, B.; Xu, J.; Wang, C.; Xiong, J. Carbon Quantum Dots–Modified Interfacial Interactions and Ion Conductivity for Enhanced High Current Density Performance in Lithium–Sulfur Batteries. *Adv. Energy Mater.* **2019**, *9*, 1802955.
- (141) Bouzas-Ramos, D.; Cigales Canga, J.; Mayo, J. C.; Sainz, R. M.; Ruiz Encinar, J.; Costa-Fernandez, J. M. Carbon Quantum Dots Codoped with Nitrogen and Lanthanides for Multimodal Imaging. *Adv. Funct. Mater.* **2019**, *29*, 1903884.
- (142) Liu, Y.; Liu, J.; Zhang, J.; Li, X.; Lin, F.; Zhou, N.; Yang, B.; Lu, L. A Brand-New Generation of Fluorescent Nano-Neural Tracers: Biotinylated Dextran Amine Conjugated Carbonized Polymer Dots. *Biomater. Sci.* **2019**, *7*, 1574–1583.
- (143) Song, Y.; Li, H.; Lu, F.; Wang, H.; Zhang, M.; Yang, J.; Huang, J.; Huang, H.; Liu, Y.; Kang, Z. Fluorescent Carbon Dots with Highly Negative Charges as a Sensitive Probe for Real-Time Monitoring of Bacterial Viability. *J. Mater. Chem. B* **2017**, *5*, 6008–6015.
- (144) Lu, F.; Song, Y.; Huang, H.; Liu, Y.; Fu, Y.; Huang, J.; Li, H.; Qu, H.; Kang, Z. Fluorescent Carbon Dots with Tunable Negative Charges for Bio-Imaging in Bacterial Viability Assessment. *Carbon* **2017**, *120*, 95–102.
- (145) Lin, F.; Li, C.; Dong, L.; Fu, D.; Chen, Z. Imaging Biofilm-Encased Microorganisms Using Carbon Dots Derived from *L. Plantarum*. *Nanoscale* **2017**, *9*, 9056–9064.
- (146) Yang, J.; Gao, G.; Zhang, X.; Ma, Y. H.; Chen, X.; Wu, F. G. One-Step Synthesized Carbon Dots with Bacterial Contact-Enhanced Fluorescence Emission Property: Fast Gram-Type Identification and Selective Gram-Positive Bacterial Inactivation. *Carbon* **2019**, *146*, 827–839.
- (147) Li, H.; Huang, J.; Song, Y.; Zhang, M.; Wang, H.; Lu, F.; Huang, H.; Liu, Y.; Dai, X.; Gu, Z.; Yang, Z.; Zhou, R.; Kang, Z. Degradable Carbon Dots with Broad-Spectrum Antibacterial Activity. *ACS Appl. Mater. Interfaces* **2018**, *10*, 26936–26946.
- (148) Wang, S.; Zhang, Y.; Zhuo, P.; Hu, Q.; Chen, Z.; Zhou, L. Identification of Eight Pathogenic Microorganisms by Single Concentration-Dependent Multicolor Carbon Dots. *J. Mater. Chem. B* **2020**, *8*, 5877–5882.
- (149) Li, W.; Zhang, H.; Zheng, Y.; Chen, S.; Liu, Y.; Zhuang, J.; Liu, W. R.; Lei, B. Multifunctional Carbon Dots for Highly Luminescent Orange-Emissive Cellulose Based Composite Phosphor Construction and Plant Tissue Imaging. *Nanoscale* **2017**, *9*, 12976–12983.
- (150) Ji, Z.; Arvapalli, D. M.; Zhang, W.; Yin, Z.; Wei, J. Nitrogen and Sulfur Co-Doped Carbon Nanodots in Living EA.Hy926 and A549 Cells: Oxidative Stress Effect and Mitochondria Targeting. *J. Mater. Sci.* **2020**, *55*, 6093–6104.
- (151) Geng, X.; Sun, Y.; Li, Z.; Yang, R.; Zhao, Y.; Guo, Y.; Xu, J.; Li, F.; Wang, Y.; Lu, S.; Qu, L. Retrosynthesis of Tunable Fluorescent Carbon Dots for Precise Long-Term Mitochondrial Tracking. *Small* **2019**, *15*, 1901517.
- (152) Liu, Y.; Liu, J.; Zhang, J.; Li, X.; Lin, F.; Zhou, N.; Yang, B.; Lu, L. Noninvasive Brain Tumor Imaging Using Red Emissive Carbonized Polymer Dots across the Blood-Brain Barrier. *ACS Omega* **2018**, *3*, 7888–7896.
- (153) Wang, L.; Wu, B.; Li, W.; Li, Z.; Zhan, J.; Geng, B.; Wang, S.; Pan, D.; Wu, M. Industrial Production of Ultra-Stable Sulfonated Graphene Quantum Dots for Golgi Apparatus Imaging. *J. Mater. Chem. B* **2017**, *5*, 5355–5361.
- (154) Li, R. S.; Gao, P. F.; Zhang, H. Z.; Zheng, L. L.; Li, C. M.; Wang, J.; Li, Y. F.; Liu, F.; Li, N.; Huang, C. Z. Chiral Nanoprobes for Targeting and Long-Term Imaging of the Golgi Apparatus. *Chem. Sci.* **2017**, *8*, 6829–6835.
- (155) Hua, X. W.; Bao, Y. W.; Wu, F. G. Fluorescent Carbon Quantum Dots with Intrinsic Nucleolus-Targeting Capability for Nucleolus Imaging and Enhanced Cytosolic and Nuclear Drug Delivery. *ACS Appl. Mater. Interfaces* **2018**, *10*, 10664–10677.
- (156) Liu, H.; Yang, J.; Li, Z.; Xiao, L.; Aryee, A. A.; Sun, Y.; Yang, R.; Meng, H.; Qu, L.; Lin, Y.; Zhang, X. Hydrogen-Bond-Induced Emission of Carbon Dots for Wash-Free Nucleus Imaging. *Anal. Chem.* **2019**, *91*, 9259–9265.

- (157) Yang, S. T.; Cao, L.; Luo, P. G.; Lu, F.; Wang, X.; Wang, H.; Mezziani, M. J.; Liu, Y.; Qi, G.; Sun, Y. P. Carbon Dots for Optical Imaging in Vivo. *J. Am. Chem. Soc.* **2009**, *131*, 11308–11309.
- (158) Xu, G.; Bao, X.; Chen, J.; Zhang, B.; Li, D.; Zhou, D.; Wang, X.; Liu, C.; Wang, Y.; Qu, S. In Vivo Tumor Photoacoustic Imaging and Photothermal Therapy Based on Supra-(Carbon Nanodots). *Adv. Healthcare Mater.* **2019**, *8*, 1800995.
- (159) Sun, S.; Chen, J.; Jiang, K.; Tang, Z.; Wang, Y.; Li, Z.; Liu, C.; Wu, A.; Lin, H. Ce6-Modified Carbon Dots for Multimodal-Imaging-Guided and Single-NIR-Laser-Triggered Photothermal/Photodynamic Synergistic Cancer Therapy by Reduced Irradiation Power. *ACS Appl. Mater. Interfaces* **2019**, *11*, 5791–5803.
- (160) Xu, W.; Chen, J.; Sun, S.; Tang, Z.; Jiang, K.; Song, L.; Wang, Y.; Liu, C.; Lin, H. Fluorescent and Photoacoustic Bifunctional Probe for the Detection of Ascorbic Acid in Biological Fluids, Living Cells and: In Vivo. *Nanoscale* **2018**, *10*, 17834–17841.
- (161) Liu, Y.; Zhi, X.; Hou, W.; Xia, F.; Zhang, J.; Li, L.; Hong, Y.; Yan, H.; Peng, C.; De La Fuentea, J. M.; Song, J.; Cui, D. Gd³⁺-Ion-Induced Carbon-Dots Self-Assembly Aggregates Loaded with a Photosensitizer for Enhanced Fluorescence/MRI Dual Imaging and Antitumor Therapy. *Nanoscale* **2018**, *10*, 19052–19063.
- (162) Zhang, M.; Zheng, T.; Sheng, B.; Wu, F.; Zhang, Q.; Wang, W.; Shen, J.; Zhou, N.; Sun, Y. Mn²⁺ Complex-Modified Polydopamine- and Dual Emissive Carbon Dots Based Nanoparticles for in Vitro and in Vivo Trimodality Fluorescent, Photothermal, and Magnetic Resonance Imaging. *Chem. Eng. J.* **2019**, *373*, 1054–1063.
- (163) Zheng, D. W.; Li, B.; Li, C. X.; Fan, J. X.; Lei, Q.; Li, C.; Xu, Z.; Zhang, X. Z. Carbon-Dot-Decorated Carbon Nitride Nanoparticles for Enhanced Photodynamic Therapy against Hypoxic Tumor via Water Splitting. *ACS Nano* **2016**, *10*, 8715–8722.
- (164) Jia, Q.; Ge, J.; Liu, W.; Zheng, X.; Chen, S.; Wen, Y.; Zhang, H.; Wang, P. A Magnetofluorescent Carbon Dot Assembly as an Acidic H₂O₂-Driven Oxygenator to Regulate Tumor Hypoxia for Simultaneous Bimodal Imaging and Enhanced Photodynamic Therapy. *Adv. Mater.* **2018**, *30*, 1706090.
- (165) Li, Y.; Zheng, X.; Zhang, X.; Liu, S.; Pei, Q.; Zheng, M.; Xie, Z. Porphyrin-Based Carbon Dots for Photodynamic Therapy of Hepatoma. *Adv. Healthcare Mater.* **2017**, *6*, 1600924.
- (166) He, H.; Zheng, X.; Liu, S.; Zheng, M.; Xie, Z.; Wang, Y.; Yu, M.; Shuai, X. Diketopyrrolopyrrole-Based Carbon Dots for Photodynamic Therapy. *Nanoscale* **2018**, *10*, 10991–10998.
- (167) Wu, F.; Chen, J.; Li, Z.; Su, H.; Leung, K. C. F.; Wang, H.; Zhu, X. Red/Near-Infrared Emissive Metalloporphyrin-Based Nanodots for Magnetic Resonance Imaging-Guided Photodynamic Therapy In Vivo. *Part. Part. Syst. Charact.* **2018**, *35*, 1800208.
- (168) Permatasari, F. A.; Fukazawa, H.; Ogi, T.; Iskandar, F.; Okuyama, K. Design of Pyrrolic-N-Rich Carbon Dots with Absorption in the First Near-Infrared Window for Photothermal Therapy. *ACS Appl. Nano Mater.* **2018**, *1*, 2368–2375.
- (169) Li, D.; Han, D.; Qu, S. N.; Liu, L.; Jing, P. T.; Zhou, D.; Ji, W. Y.; Wang, X. Y.; Zhang, T. F.; Shen, D. Z. Supra-(Carbon Nanodots) with a Strong Visible to near-Infrared Absorption Band and Efficient Photothermal Conversion. *Light: Sci. Appl.* **2016**, *5*, 16120.
- (170) Song, J.; Yang, X.; Yang, Z.; Lin, L.; Liu, Y.; Zhou, Z.; Shen, Z.; Yu, G.; Dai, Y.; Jacobson, O.; Munasinghe, J.; Yung, B.; Teng, G.-J.; Chen, X. Rational Design of Branched Nanoporous Gold Nanoshells with Enhanced Physico-Optical Properties for Optical Imaging and Cancer Therapy. *ACS Nano* **2017**, *11*, 6102–6113.
- (171) Zhao, S.; Wu, S.; Jia, Q.; Huang, L.; Lan, M.; Wang, P. Lysosome-Targetable Carbon Dots for Highly Efficient Photothermal/Photodynamic Synergistic Cancer Therapy and Photoacoustic/Two-Photon Excited Fluorescence Imaging. *Chem. Eng. J.* **2020**, *388*, 124212.
- (172) Yang, W.; Wei, B.; Yang, Z.; Sheng, L. Facile Synthesis of Novel Carbon-Dots/Hemin Nanoplateforms for Synergistic Photothermal and Photo-Dynamic Therapies. *J. Inorg. Biochem.* **2019**, *193*, 166–172.
- (173) Li, J.; Yang, S.; Deng, Y.; Chai, P.; Yang, Y.; He, X.; Xie, X. Emancipating Target-Functionalized Carbon Dots from Autophagy Vesicles for a Novel Visualized Tumor Therapy. *Adv. Funct. Mater.* **2018**, *28*, 1800881.
- (174) Sung, S.; Su, Y.; Cheng, W.; Hu, P.; Chiang, C.; Chen, W.; Hu, S. Graphene Quantum Dots-Mediated Theranostic Penetrative Delivery of Drug and Photolytics in Deep Tumors by Targeted Biomimetic Nanosponges. *Nano Lett.* **2019**, *19*, 69–81.
- (175) Li, S.; Su, W.; Wu, H.; Yuan, T.; Yuan, C.; Liu, J.; Deng, G.; Gao, X.; Chen, Z.; Bao, Y.; Yuan, F.; Zhou, S.; Tan, H.; Li, Y.; Li, X.; Fan, L.; Zhu, J.; Chen, A. T.; Liu, F.; Zhou, Y.; Li, M.; Zhai, X.; Zhou, J. Targeted Tumour Theranostics in Mice via Carbon Quantum Dots Structurally Mimicking Large Amino Acids. *Nat. Biomed. Eng.* **2020**, *4*, 704–716.
- (176) Gao, P.; Liu, S.; Su, Y.; Zheng, M.; Xie, Z. Fluorine-Doped Carbon Dots with Intrinsic Nucleus-Targeting Ability for Drug and Dye Delivery. *Bioconjugate Chem.* **2020**, *31*, 646–655.
- (177) Scialabba, C.; Sciortino, A.; Messina, F.; Buscarino, G.; Cannas, M.; Roscigno, G.; Condorelli, G.; Cavallaro, G.; Giammona, G.; Mauro, N. Highly Homogeneous Biotinylated Carbon Nanodots: Red-Emitting Nanoheaters as Theranostic Agents toward Precision Cancer Medicine. *ACS Appl. Mater. Interfaces* **2019**, *11*, 19854–19866.
- (178) Ghosh, S.; Ghosal, K.; Mohammad, S. A.; Sarkar, K. Dendrimer Functionalized Carbon Quantum Dot for Selective Detection of Breast Cancer and Gene Therapy. *Chem. Eng. J.* **2019**, *373*, 468–484.
- (179) Han, J.; Na, K. Journal of Industrial and Engineering Chemistry Transfection of the TRAIL Gene into Human Mesenchymal Stem Cells Using Biocompatible Polyethyleneimine Carbon Dots for Cancer Gene Therapy. *J. Ind. Eng. Chem.* **2019**, *80*, 722–728.
- (180) Dunbar, C. E.; High, K. A.; Joung, J. K.; Kohn, D. B.; Ozawa, K.; Sadelain, M. Gene Therapy Comes of Age. *Science (Washington, DC, U. S.)* **2018**, *359*, 175.
- (181) Li, P.; Liu, S.; Cao, W.; Zhang, G.; Yang, X.; Gong, X.; Xing, X. Low-Toxicity Carbon Quantum Dots Derived from Gentamicin Sulfate to Combat Antibiotic Resistance and Eradicate Mature Biofilms †. *Chem. Commun.* **2020**, *56*, 2316–2319.
- (182) Li, P.; Han, F.; Cao, W.; Zhang, G.; Li, J.; Zhou, J.; Gong, X.; Turnbull, G.; Shu, W.; Xia, L.; Fang, B.; Xing, X.; Li, B. Carbon Quantum Dots Derived from Lysine and Arginine Simultaneously Scavenge Bacteria and Promote Tissue Repair. *Appl. Mater. Today* **2020**, *19*, 100601.
- (183) Zhu, C.; Li, H.; Wang, H.; Yao, B.; Huang, H.; Liu, Y.; Kang, Z. Negatively Charged Carbon Nanodots with Bacteria Resistance Ability for High-Performance Antibiofilm Formation and Anticorrosion Coating Design. *Small* **2019**, *15*, 1900007.
- (184) Lu, S.; Liu, L.; Wang, H.; Zhao, W.; Li, Z.; Qu, Z.; Li, J.; Sun, T.; Wang, T.; Sui, G. Biomaterials Science Carbon Dots for Bioimaging and Antitumor. *Biomater. Sci.* **2019**, *7*, 3258–3265.
- (185) Zhang, F.; Zhang, M.; Zheng, X.; Tao, S.; Zhang, Z.; Sun, M.; Song, Y.; Zhang, J.; Shao, D.; He, K.; Li, J.; Yang, B.; Chen, L. RSC Advances Cancer Theranostics †. *RSC Adv.* **2018**, *8*, 1168–1173.
- (186) Lin, C.; Chang, L.; Chu, H.; Lin, H.; Chang, P.; Wang, R. Y. L.; Unnikrishnan, B.; Mao, J.; Chen, S. High Amplification of the Antiviral Activity of Curcumin through Transformation into Carbon Quantum Dots. *Small* **2019**, *15*, 1902641.
- (187) Tong, T.; Hu, H.; Zhou, J.; Deng, S.; Zhang, X.; Tang, W. Glycyrrhizic-Acid-Based Carbon Dots with High Antiviral Activity by Multisite Inhibition Mechanisms. *Small* **2020**, *16*, 1906206.
- (188) Chen, R.; Liu, G.; Sun, X.; Cao, X.; He, W.; Lin, X.; Liu, Q.; Zhao, J.; Pang, Y.; Li, B.; Qin, A. Dots Suppress Osteoclastic Osteolysis Via. *Nanoscale* **2020**, *12*, 16229–16244.
- (189) Wang, H.; Xie, Y.; Na, X.; Bi, J.; Liu, S. Fluorescent Carbon Dots in Baked Lamb: Formation, Cytotoxicity and Scavenging Capability to Free Radicals. *Food Chem.* **2019**, *286*, 405–412.
- (190) Wei, X.; Li, L.; Liu, J.; Yu, L.; Li, H.; Cheng, F.; Yi, X.; He, J.; Li, B. Green Synthesis of Fluorescent Carbon Dots from Gynostemma for Bioimaging and Antioxidant in Zebra Fi Sh. *ACS Appl. Mater. Interfaces* **2019**, *11*, 9832–9840.